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ENVIRONMENTAL HEALTH SERIES
Radiological Health

COMMON LABORATORY INSTRUMENTS FOR MEASUREMENT OF RADIOACTIVITY

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
Consumer Protection and Environmental Health Service
Environmental Control Administration

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COMMON LABORATORY INSTRUMENTS FOR MEASUREMENT OF RADIOACTIVITY

April 1967

Interlaboratory Technical Advisory Committee

Report No. 2

by the
Subcommittee on Physics and Instrumentation

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Mr. Robert W. Coulter
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Foreword

This is the second report issued under the auspices of the Interlaboratory Technical Advisory Committee (ITAC). The ITAC advised the Center Director, National Center for Radiological Health, on technical problems of mutual concern to the various laboratories within the Center. The first report, sponsored by the ITAC of the former Division of Radiological Health, was entitled "Routine Surveillance of Radioactivity around Nuclear Facilities."

Report No. 2 was prepared by the ITAC Subcommittee on Physics and Instrumentation. It was written to clarify requirements and characteristics of instrumentation used to measure levels of ionizing radiation experienced by man in his environment, and to assist responsible radiological health agencies in selecting optimum surveillance and counting systems.

The members of the Subcommittee on Physics and Instrumentation, and particularly Mr. D. Edward Rushing, who served as Chairman, are commended for their efforts in preparing this report. The document is a valuable addition to the libraries of persons engaged in radiation protection.

James G Terrill

James G. Terrill, Jr.
Center Director
National Center for Radiological Health

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Abstract

This ~~second~~ report, issued under the auspices of the Interlaboratory Technical Advisory Committee (ITAC), was prepared by the Subcommittee on Physics and Instrumentation. It clarifies requirements and characteristics of instrumentation used to measure levels of ionizing radiation experienced by man in his environment, and assists responsible radiological health agencies in selecting optimum surveillance and counting systems.

Representative products and manufacturers are named for identification only, and listing does not imply endorsement by the Public Health Service and the U.S. Department of Health, Education, and Welfare.

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[1] The Division of Radiological Health was reorganized January 1, 1967. It is now the National Center for Radiological Health, with Mr. James G. Terrill, Jr., as Center Director.

COMMON LABORATORY INSTRUMENTS FOR MEASUREMENT OF RADIOACTIVITY

Introduction

Donald G. Remark [1]

The Public Health Service has been performing radiological analyses on environmental samples for many years. The continuing surveillance of environmental radiation levels is necessary to determine when radioactivity values increase to levels demanding appropriate Federal or State action. The Public Health Service has also been given the responsibility for the offsite environmental survey programs in direct support of Continental and Pacific nuclear test operations.

The factors influencing the selection of the instrumentation for the determination of low levels of radioactivity in the environment are described. To determine the effects of nuclear testing, isotope research, and reactor operations on the environment, it is necessary to have a knowledge of existing activity levels contributed by these operations, as well as the background from natural sources.

Some aspects of nuclear instrumentation system characteristics and requirements are clarified, and it is not intended that detailed specifications for purchase orders be included. Apparatus and instruments for the radiochemical preparation of samples to be counted are not included, nor is the relative ease of making the sample into a form suitable for the particular instrument considered. Although an initial purchase of a costlier or different type of system may lead to substantial savings in the cost of sample preparation and time, such factors have not been included except as general comments. The guidelines are intended to enable the purchaser to evaluate manufacturers' proposals and to assist in the selection of optimum systems for individual radiological health agencies or activities. This report is divided into three chapters: alpha-counting systems, beta-counting systems, and gamma-counting systems. Each chapter includes pertinent restrictions on equipment based on experience gained by personnel actually involved in determination of low levels of radioactivity. The chapters as written are complete, and some basic considerations are repeated.

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[1] Staff Officer, Technical Services Branch, National Center for Radiological Health, PHS, USDHEW, Rockville, Md. 20852.

The instrumentation and the chemical procedures used to identify submicroquantities of radionuclides in the presence of macroquantities of nonradioactive material require a high degree of development to achieve the maximum sensitivity necessary for quantitative data. The selection and purchase of these instruments require consideration of many facts, including—but not limited to—performance characteristics, initial and maintenance costs, compatibility with existing systems, capacity for expansion, and availability of professional and technical personnel. The ability to expand economically and also the ability to analyze nonroutine samples will depend on the careful selection of basic instrumentation. Preamplifiers, amplifiers, scalers, timers, high-voltage power supplies, voltage regulators, and appropriate detectors are examples of basic instruments common to radiation surveillance laboratories. Initially the small laboratory should invest the major portion of its funds in these basic instruments. Purchasing will be dictated by initial and projected laboratory workloads, local conditions, and available funds. In order to develop initial laboratory capabilities, these instruments should be interchangeable with units of the same type and be capable of use in a variety of different systems. An alternative to the unit instrument approach is the "Standard Nuclear Instrument Modules" as adopted by the U.S. Atomic Energy Commission. These modules, referred to as AEC Standard, allow greater flexibility in setting up various system configurations.

Versatility of counting systems will be increased if the equipment is supplied by the same manufacturer. The manufacturer should guarantee the proper operation of the equipment for at least 1 year. Equipment downtime will be shortened and maintenance will be simplified if the manufacturer maintains a local repair service, and if the equipment is constructed with plug-in circuit boards. Transistorized equipment generally is more reliable than the vacuum tube equipment. Long-term stability and reliability of all counting equipment will be improved when the counting laboratory has a filtered air supply, and the temperature and humidity controls are separate from the rest of the building. The temperature and humidity of the air supplied to the counting rooms should be maintained within narrow ranges. The required capacity of the air-conditioning system will depend on the dissipation of heat in areas of counting instruments, concentrations of personnel, and the power supply requirements of the instrumentation used. Freedom from gross excursions, transients, and failures is an important restriction on power lines and may necessitate the installation of protective circuitry for the counting systems. One form of protective circuitry, used when gross excursions of power lines are a possibility, is a manual reset control. This is a desirable feature on voltage regulators and power supplies.

Gross alpha and gross beta counting may be performed with a single system consisting of a shielded gas proportional counter chamber, high-voltage power supply, amplifier, scaler, and timer. This basic system may be modified so it is capable of gross gramma counting by replacing the counting chamber with a NaI(Tl) crystal, photomultiplier tube, and background reduction shield. The addition of a linear amplier and single-channel analyzer to this system will provide gamma energy identification and counting capability. Additional modifications to these systems may take the form of additional shielding, guard chambers, coincidence circuitry, and automatic sample changers, depending on sample load. Further desirable and major acquisitions will be a multichannel-analyzer system, a shield, and a large NaI(Tl) crystal. Only after a laboratory has several interchangeable systems should new systems be purchased for the purpose of performing one or two specialized counting operations. As the laboratory becomes more sophisticated, highly refined equipment may be needed, such as multiparameter analyzers and whole-body counters. These instruments, as well as developments in solid state detectors, are not encompassed in this report.

COUNTING EQUIPMENT

Scalers, timers, high-voltage power supplies, and voltage regulators are examples of basic counting equipment commonly used in the systems mentioned herein. An excellent reference to the manufacturers of these and other instruments is the "Nucleonics Buyers Guide" published by McGraw-Hill, Inc.

Scaler

The scaler is one of the more common instruments found in a counting facility. Specifications with respect to the number and type of decades, minimum sensitivity, maximum counting rate, pulse-pair resolution, preset count capability, and maximum pulse height without double pulsing will adequately distinguish between most scalers on the market. A scaler should have a pulse-pair resolving time of between 1 and 5 microseconds and be capable of counting at a 200 kiloHertz. An adjustable discriminator with a minimum sensitivity of 25 millivolts and the capability of accepting 10-volt pulses without double pulsing are desirable features. The ability of a scaler to control other instruments after a preset count is reached is also desirable. The preset count should be selectable to any digit, and the scaler should have a capacity of 99,999 without electromechanical registers. Another desirable feature is a built-in pulse generator, which can be used to check the operation of the scaler.

Timer

Two basic methods are available for obtaining a time base for radiological determinations. The selection of the method to be used is one of the more important considerations in differentiating between timers. The accuracy of the time base is directly proportional to the maximum accuracy of the time measurement. One method of obtaining a time base depends on the line frequency supplied to the installation. If this line frequency is constant at 60 counts per second, the timer obtaining the time base from this line frequency will be accurate; however, this is not always the case. The second method of obtaining a time base, by means of a crystal-controlled oscillator, is more expensive, but it is capable of extreme accuracy and is to be preferred in those locations where the accuracy and stability of the line frequency are not assured.

The ability to control several pieces of equipment by means of open or closed relay contacts, after a preset time interval is reached, is another desirable feature. This timer should be capable of being preset to any time interval between 1 and 1,000 minute increments.

Voltage Regulators

There are three general types of voltage regulators: the constant voltage transformer, electromechanical variable autotransformer, and electronic bridge. Because the instrumentation used in the laboratories may be extremely sensitive to electronic noise and voltage variation, the electronic bridge regulator is recommended because of its faster response. It will be more economical, if laboratory capabilities are to be expanded, to obtain a larger unit than is initially necessary. The larger capacity unit can be obtained at a small increase in cost.

High-Voltage Power Supply

For maximum versatility and interchangeability, a high-voltage d.c. power supply, to be used for gas-flow and scintillation counting, should be capable of supplying any voltage in the range of 500 to 5,000 volts d.c. A voltage supply with a voltage output range of 500 to 3,000 volts d.c. will be sufficient if the counting equipment uses gas mixtures such as 90 percent argon and 10 percent methane, or 96 percent helium and 4 percent isobutane, rather than methane. This power supply should be capable of supplying at least 1 milliampere at maximum voltage. Since larger current capabilities can be obtained for small increases in cost, it might be more economical initially if a unit is purchased with output current capabilities in excess of 1 milliampere so more than one piece of equipment can be operated from a central supply by means of appropriate voltage dividers.

Electronic noise and ripple, a component of the supply's output voltage, may cause spurious counts in a counting system. The exact noise level causing spurious counts is a function of the system's parameters. A large number of power supplies are available with noise and ripple values below 80 millivolts, peak-to-peak, and these can be used with the systems described. Power supplies having noise and ripple values larger than 80 millivolts, peak-to-peak, may not cause spurious counts, but this should be checked before purchasing.

This same caution should be observed before purchasing a unit that has a stability value in excess of 0.1 percent change in output voltage per day. This is because counting reproducibility is a function of the stability of the high-voltage supply.

Chapter I. Alpha-Counting Instruments

ALPHA PROPORTIONAL COUNTERS

Dr. S. David Shearer[1]

Principles of Proportional Counters

The detection of nuclear radiation is based on the ionizing properties of radiation. Such ionization produces charged particles or ions from neutral atoms and molecules. Basically, radiation detection instruments vary only in the medium in which the ionization takes place and the manner in which ionization is identified and detected. The majority of laboratory instruments used for the detection of alpha and beta radiation are based on the principle of gas ionization by radiation.

A review of the basic differences between ionization chambers, proportional counters, and Geiger-Mueller counters is appropriate before methods of detection of radiation by alpha proportional counters are presented. This can best be accomplished by considering the regions of instrument response first.

Regions of Instrument Response

If two collecting electrodes are placed in an enclosed gas volume containing a source of radiation, and if a source of d.c. voltage is impressed across these electrodes, it will be observed that five regions of instrument response will occur as the voltage is increased above zero. By plotting the relative number of ion

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pairs collected (or pulse height) versus applied voltage, a general curve results (fig. 1). The five regions of instrument response are noted on this figure and are briefly discussed below.

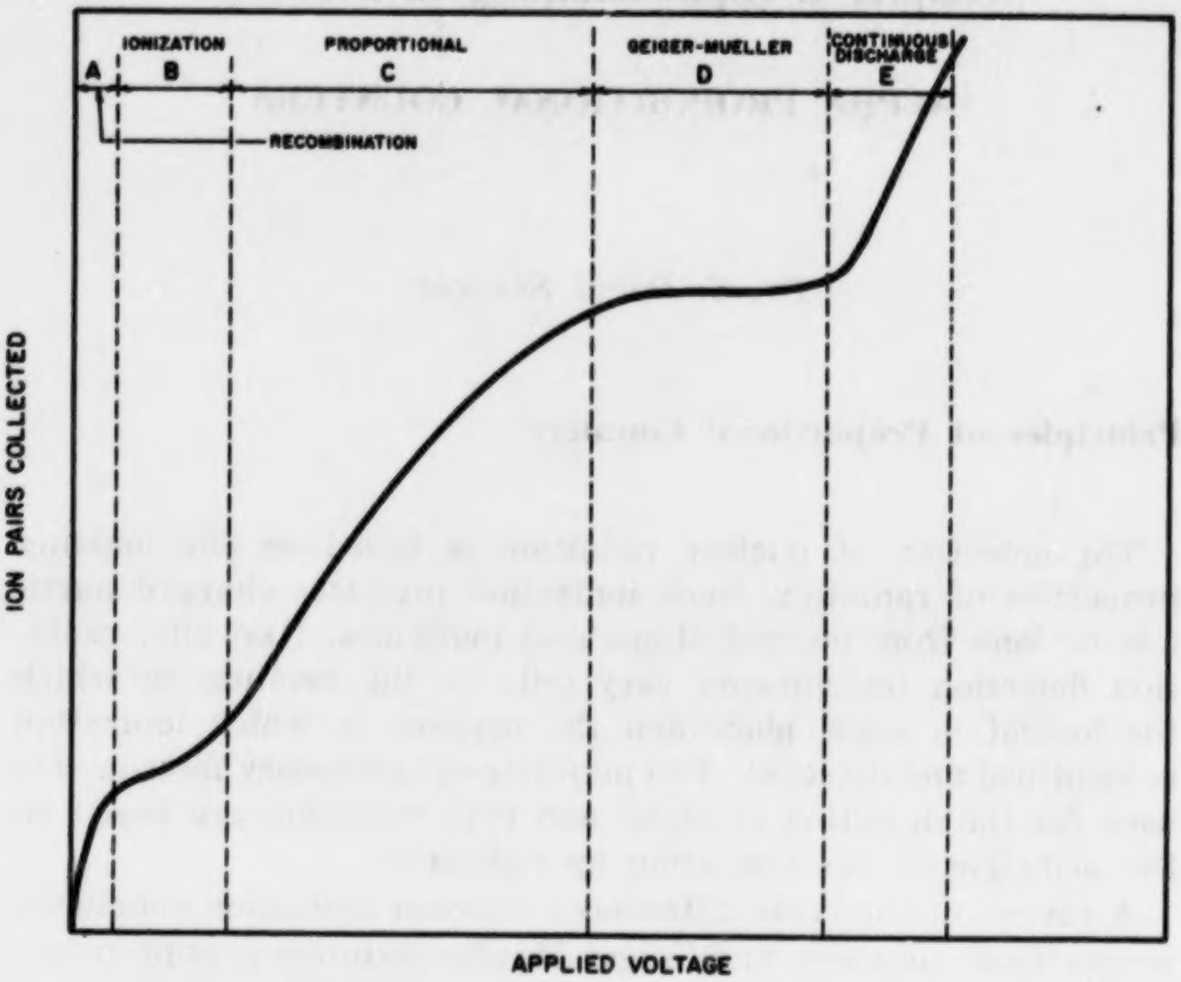


Figure 1. Regions of instrument response.

Ion Recombination Region

In this region as the voltage is increased slightly above zero, the voltage gradient between the two electrodes is not sufficient to cause the ions produced by the radiation to have a significant velocity of passage through the gas medium; consequently some of the ion pairs formed will not reach the collector electrodes but will recombine with each other. This recombination decreases as the voltage is increased.

Ionization Chamber Region

The second region of instrument response begins at the voltage point where a negligible number of ion pairs recombine with each other; that is, the point where initially all the ion pairs formed by the radiation are collected by the collector electrodes. At this point, the voltage gradient is sufficient to accelerate the negative member of the ion pair toward the anode and the positive ion toward the cathode. In this region a saturation flow of ions occurs. The flow of ions is equal to the number of ions produced by the radiation entering the chamber; thus ions are collected in a ratio of 1 to 1 with those formed.

Ionization chambers are normally operated in this voltage area, and it is the first of three regions of operation of gas ionization instruments.

Proportional Counter Region

When the voltage is increased above the region of saturation current flow, it will be observed that more ions are collected at the electrode than are formed by the radiation source. This is because the voltage gradient between the electrodes is sufficiently high to accelerate the ions so that they acquire enough energy to cause secondary ionization in the filling gas. Such secondary ionization results in an amplification of the primary ion current produced by the radiation. This amplification is referred to as the "gas-amplification factor," and is defined as the total number of ion pairs produced by a single original ion pair. The gas-amplification factor is unity in the ionization chamber region, but it can be as large as 10^4 or more in the proportional region. When the proportional region is reached, the size of the pulse is a function of the voltage impressed between the electrodes, and it is proportional to the number of primary ion pairs formed. Because of differences in total ionization within the chamber, alpha particles produce larger pulses of current than do beta particles of the same energy. This region of proportionality between pulse size and particle energy is the region where alpha and beta proportional counters are operated.

Geiger-Mueller Region

Further increases in gas amplification occur by increasing the voltage above the proportional region. Eventually a point is reached where an avalanche of electrons to the anode occurs. In this region, the pulse amplitude produced is essentially independent of the total ionization of the event. This is called the Geiger-Mueller region.

Continuous Discharge Region

If the voltage is increased beyond the Geiger-Mueller region, an arc is formed and a continuous discharge occurs. If an instrument is operated in this region even for a few seconds, the electrodes will be seriously damaged.

The following comments can be made regarding instruments operating in Regions B, C, and D of figure 1. Ionization counters operated in Region B have the advantage of being simple, dependable, and rugged. They have the disadvantage of supplying only a very small pulse, and therefore require a large amount of external amplification for pulse-type operation. Proportional counters operated in Region C, particularly internal proportional counters, will be discussed in greater detail in subsequent sections of this chapter. Geiger-Mueller counters operated in Region D have a high sensitivity to all different types of radiation, and because of the large size of the output pulse they require much less external amplification. This reduces the complexity and cost of auxiliary electronic equipment. Geiger-Mueller detector tubes are relatively simple, and a great deal of design flexibility can be utilized. Other details regarding Geiger-Mueller counters are discussed in a separate chapter.

CHARACTERISTICS AND USE OF ALPHA PROPORTIONAL COUNTERS

Radiation detection instruments operated in the proportional region of figure 1 (proportional counters) offer several inherent

advantages over Geiger-Mueller counters. The first is the shorter resolving time (the minimum time interval between two distinct events that will permit both to be counted) of proportional counters. The resolving time of proportional counters is on the order of 10 microseconds (although it can be as low as 1 microsecond). This compares with a longer resolving time for most Geiger-Mueller counters. This means that much higher count rates may be handled by a proportional counter.

More amplification is needed than for a Geiger-Mueller counter because of the nature of pulses in a proportional counter. Proportional counters require better voltage regulation than Geiger-Mueller counters. Because of the ability to vary voltage discrimination, proportional counters can discriminate between alpha and beta pulses whereas Geiger-Mueller counters cannot.

Proportional counters usually are accepted as the instrument of choice for both gross alpha and gross beta radioactivity detection because of these characteristics. Commercially available proportional counters usually employ a gas-flow system and are operated either as a windowless-type counter or a window-type counter. The counting gas is usually one of the noble gases mixed with a small amount of polyatomic gas. The polyatomic gas is used to improve the characteristics of the counting gas. The gas mixture most widely used for proportional counting is 90 percent argon and 10 percent methane (P-10 gas).

Counting Chamber Construction

Many internal proportional counters use a steel hemisphere as a counting chamber. The bottom of the chamber supports the samples and is generally of a drawer-type construction. It can be pulled out for insertion of the sample and pushed in for counting operations. In the case of windowless-type internal proportional counters, there is an "O" ring seal between the top and bottom portions of the counting chamber. The counting chamber serves as the cathode in the electronic circuitry. The collector electrode (the anode) is in most cases a very small diameter wire (usually tungsten) in the shape of a small loop.

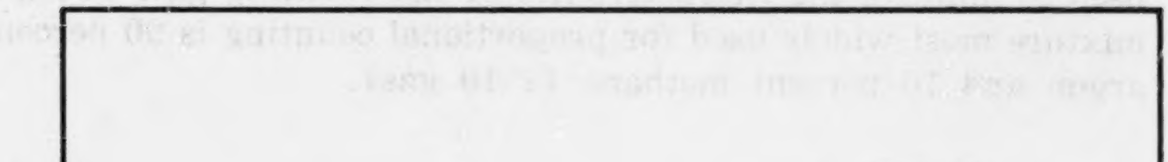
The gas purging features of proportional counters consist of a continuous flow-through system with an inlet and outlet port. The counting gas, after passing through the chamber, is normally passed through a small "bubbler." By observing this "bubbler" one is able to see the gas-flow rate through the chamber and changes can be made, if necessary. The flow rate has a slight positive pressure that is maintained inside the chamber to insure against air leaks. This is particularly true in the case of windowless-type counters. In these counters, referred to as internal proportional counters, the samples are positioned inside the hemispherical chamber.

In the window-type proportional counters, the sample is positioned outside the counting chamber and the radioactive particles being counted pass through an ultra-thin window.

Detector Operation

Proportional counters have a characteristic voltage-count rate curve with two distinct counting plateaus. Figure 2 presents a typical voltage-count rate curve for a proportional counter, and the two distinct counting plateaus are shown.

The first plateau is known as the "alpha plateau" and occurs between voltage settings of 900 to 1,300 volts with P-10 gas.



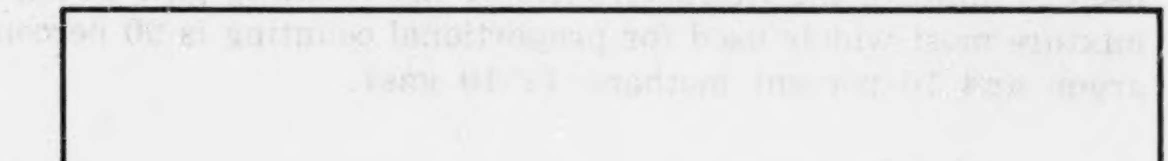


Figure 2. Proportional counter characteristic curve.

The second plateau, known as the "beta plateau," occurs between voltage settings of 1,600 and 2,000 volts. The length of the voltage plateau will depend on the particular geometry of the counting chamber. At voltage settings on the alpha plateau, beta particles are not counted because of their lower energy and smaller total ionization of the counting gas. This factor is extremely important and accounts for the widespread use of proportional counters for gross alpha and gross beta radioactivity measurements.

The most common procedure in proportional counter operation is to determine first the alpha radioactivity at the plateau setting, then change the voltage settings to the beta plateau, and count both alpha and beta radioactivity. The alpha count is then subtracted from the "alpha plus beta" count to arrive at the beta count. This practice is not theoretically correct because alpha plateaus usually exhibit a slope of about 1 percent per hundred volts. This slope results in the alpha count rate being higher on the beta plateau than on the alpha plateau. For extremely accurate beta assay work, the slope of the alpha plateau can be measured, and a correction can be made for the effect of the alpha count rate on the beta count rate. This condition is likely to manifest itself only when counting is done at very high voltages on the beta plateau. For routine environmental radioassay work, where the alpha radioactivity is much less than the beta radioactivity, it is not necessary to make such corrections.

In addition to the detector, a proportional counter includes several other components. The usual practice is to purchase a proportional counting system containing the detector, a stable high-voltage supply, an amplier, a scaler, and a timer.

Window-Type Alpha Proportional Counters

There are several commercially available alpha proportional counters that are operated as window-type counters. Present commercial window-type instruments have the optional capability of simultaneously recording alpha and beta radioactivity. This is accomplished by electronically discriminating the low-voltage beta pulses from the higher alpha pulses.

One of the problems in simultaneously detecting alpha and beta particles with such instruments arises from large amounts of solids being present on the counting planchets. If appreciable solids are present to degrade the energy of the alpha particle, the resulting particles of degraded energy are counted as beta particles, not alpha particles; for example, with large amounts of solids present the apparent beta radioactivity may be considerably higher because of partially absorbed alpha particles. Also, alpha particles passing through the window of such instruments may have their energy partially degraded and will be counted as beta radioactivity. The self-absorption of alpha particles in the sample solids is a problem encountered with practically all types of counting systems. In window-type systems it adds to other problems as described below.

When using window-type alpha proportional counters, the sample should be mounted close enough to the thin window so that the alpha particles can pass through the air and the window without being completely attenuated. It is also important that the thin windows do not become broken or contaminated.

There are several distinct advantages to window-type proportional counters, and these types are more widely used. These advantages are (1) less contamination of the counting chamber, (2) simpler techniques of sample preparation, (3) less effects of moisture on counting gas contamination, and (4) fewer counting problems associated with conductivity effects.

Windowless-Type Alpha Proportional Counters

The windowless-type counter is widely used and is referred to as an "internal proportional counter." In this type of counter the sample is placed inside the counting chamber. Since there is no attenuation of the alpha particles by air or the window, the detection of lower energy alpha particles is possible. The chamber is operated at a slight positive pressure created by the continuous flow gas system.

The internal proportional counters also have a higher counting efficiency than window-type counters. Such counters have 50 percent or 2π geometry; that is, 50 percent of the total number of particles emitted by the source enter the counting chamber. These factors are discussed below.

There are many factors that affect alpha internal proportional counting. These include background, plateaus, counting efficiency, planchets, purging gas, sample preparation, and instrument rooms.

Background

Alpha background generally obtained with internal proportional counters should be less than 0.1 count per minute. Backgrounds signicantly greater than this will be obtained if there is contamination. Such contamination can arise from dirty counting chambers, sample solids, or from the materials of construction of the counting chamber. The latter source is not a serious problem in currently available commercial proportional counters.

In any type of proportional counting, backgrounds should be checked regularly and be made a part of the record. Depending on sample load and other considerations, it is good practice to determine instrument backgrounds at the beginning of each day, in the middle of the day, and in the late afternoon. Some laboratories make a practice of determining backgrounds after every two to four sample counts. It is important in routine operations to check backgrounds with empty planchets of the same type as are being used for unknown samples. It is also a good idea to use one set of planchets out of each new batch purchased as background planchets. These should be kept separate from planchets that contain unknown samples.

Plateaus

The alpha plateaus for internal proportional counters generally extend from about 900 to 1,300 volts with 90 percent argon and 10 percent methane. They are usually flat and have a slope of about 1 percent per 100 volts. The operating voltage for alpha counting is estimated to be in the middle one-third of the plateau. In any proportional counting, it is important to know the shape of the plateau and how the shape of this plateau may vary with changing operating conditions.

It is common operating procedure to do the counting on the flattest portion of the alpha plateau. This may not be the best place to count. If planchets containing unknown samples contain appreciable amounts of solids, these solids cause energy degradation of the alpha particles, and it may be desirable to count farther up on the alpha plateau. By following this procedure, the alpha particles that have had a portion of their energy degraded in passing through the planchet solids still have sufficient energy left to cause ionization and be counted. Samples with low solids content can be counted at any point on the alpha plateau without serious error.

It is common for alpha counting to be done just slightly above the knee of the alpha plateau. This is considered a less effective procedure because a decrease in amplication will cause decreased count rates.

It is also a poor practice to obtain alpha plateaus with an essentially weightless standard source, select an operating voltage from the plateau so determined, and then count samples with varying thicknesses of solids. It should be stressed that alpha plateaus (or beta plateaus) should be obtained with approximately the same quantity of solids on the planchet as will be present when unknown samples are counted.

It has also been found that plateaus obtained with an extended radiation source will be narrower in voltage range and have a higher operating voltage than plateaus that are determined with a point source. In order to illustrate this effect, figure 3 shows alpha plateau determinations using a radium D-E-F point source and three different weights of an extended U_3O_8 source in 2-inch stainless steel planchets. It is evident from this figure that an operating voltage of 1,075 would be selected using the radium D-E-F source, whereas an operating voltage of 1,175 would be selected using the extended source. The latter voltage would be the proper operating voltage if unknown samples are routinely being counted in 2-inch planchets.

Another point which should be mentioned is that of the multiplicative factor of alpha counts on the beta plateau. If an alpha source is counted on the alpha plateau and then counted on the beta plateau, more counts will be obtained on the beta plateau. This increase is on the order of 5 percent for a substantial number of samples. The reasons for this increase will be discussed in more detail in the chapter on beta proportional counting. If appreciable solids are present on the planchets, there will be energy degradation of the alpha particles passing through the solids. The particles that escape into the counting chamber with a degraded energy will not have sufficient energy to cause proper secondary ionization of the gas on the alpha plateaus. Because of the higher voltage gradients existing on the beta plateau, these degraded energy alpha particles will be counted. Also, for large diameter extended sources, alpha particles near the periphery of the sources will escape into a relatively insensitive volume of the counting chamber, and for the reason just cited will not be counted on the alpha plateau but will be counted on the beta plateau.

When alpha counts are subtracted from "alpha plus beta" counts to obtain the true beta counts, the increased efficiency of alpha

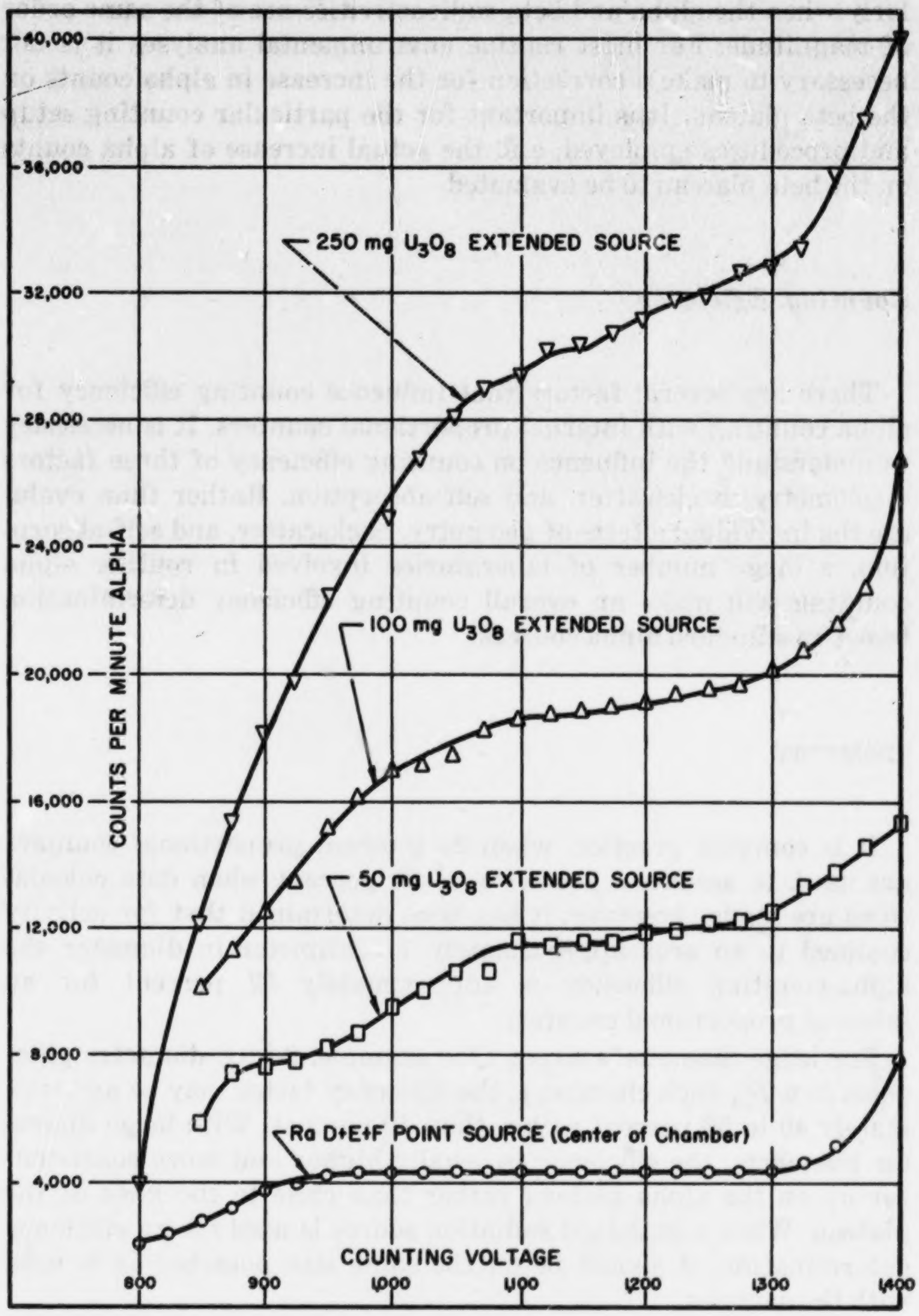


Figure 3. Alpha plateaus—Nuclear Measurements Corp. proportional counter Model PC-3A, 2-inch-diameter stainless steel planchets.

counting on the beta plateau can lead to significant error, particularly when the alpha and beta radioactivities are of the same order of magnitude. For most routine environmental analyses it is not necessary to make a correction for the increase in alpha counts on the beta plateau. It is important for the particular counting setup and procedures employed, and the actual increase of alpha counts on the beta plateau to be evaluated.

Counting Efficiency

There are several factors that influence counting efficiency for alpha counting with internal proportional counters. It is necessary to understand the influence on counting efficiency of three factors —geometry, backscatter, and self-absorption. Rather than evaluate the individual effects of geometry, backscatter, and self-absorption, a large number of laboratories involved in routine alpha counting will make an overall counting efficiency determination based on standard alpha sources.

GEOMETRY

It is common practice, when 2π internal proportional counters are used, to assume a geometry of 50 percent when data calculations are made; however, it has been determined that for activity confined to an area approximately 1 centimeter in diameter the alpha-counting efficiency is approximately 52 percent for an internal proportional counter.

For large diameter sources (for example, 2-inch-diameter planchets in a 2¼-inch chamber), the efficiency factor may be approximately 40 to 50 percent rather than 50 percent. With large diameter planchets, the efficiency is usually higher and more consistent far up on the alpha plateau rather than close to the knee of the plateau. When a standard radiation source is used for an efficiency determination, it should be on the same size planchet as is used with the samples.

For 2-inch hemispherical proportional counting chambers, the maximum efficiency for counting is obtained by the use of planchets, which are approximately 1 centimeter in diameter. It is more practical in most laboratories to use planchets 2 inches in diameter. When these are used, the efficiency figures should be carefully checked, and textbook values should not be used.

BACKSCATTER

The alpha backscatter factor varies with the type of planchet, increasing with increases in the atomic number of the planchet material. The increase in count rate because of alpha backscatter is on the order of 2 percent when stainless steel planchets are used.

SELF-ABSORPTION

This is an extremely important factor and is perhaps the largest source of error in alpha analysis. When more than a few milligrams of solids are present on a 2-inch planchet, self-absorption corrections become necessary.

It is common procedure to determine self-absorption corrections on a purely empirical basis by adding known amounts of radioactive material to different weights of suitable inorganic solids. This is satisfactory when the unknown solids being counted approximate the character of the carrier solids and contain alpha particles with approximately the same energy as the standard alpha source. This is seldom the case, however, particularly when a wide variety of environmental media are being analyzed for alpha radioactivity.

No universal procedures can be given for self-absorption corrections. Each laboratory employs the procedures it deems best for its particular work.

Self-absorption correction factors should not be treated lightly and should be determined by each laboratory for the particular type of samples being analyzed. Handbook self-absorption corrections determined by other laboratories should not be used without verifying that they apply to the particular samples being analyzed.

When standards are used for instrument calibration or for determination of counter efficiency they should be prepared very carefully. Preferably the carrier sample solids should be of the same type as unknown samples, and thickness should also be the same as unknown samples.

The above discussion has cited some of the factors that should be considered in the corrections for counting efficiency in alpha analysis. Careful techniques and attention to details will result in the ability to perform good alpha analyses.

Counting Planchets

A wide variety of planchets is available, and it is not too important which type of planchet is used as long as certain factors are realized.

Natural alpha radioactivity present in the materials from which planchets are made shall be considered. With presently available commercial planchets, background effects are not serious but should be checked.

There should be adequate electrical conductivity between the planchet and the support upon which it rests. In some commercially available instruments, it may be necessary to mill off a very thin layer of the anodized aluminum sample holder in order to provide good electrical conductivity.

It is impractical to reuse planchets; therefore this is not recommended. When reuse becomes necessary, stainless steel planchets have been most satisfactory. Reused planchets should always be recounted to establish the degree of cleanliness.

Counting Gas

The most common gas used for alpha proportional counting is 90 percent argon and 10 percent methane. Methane alone can be used satisfactorily as a counting gas. A gaseous mixture of 4 percent isobutane and 96 percent helium is quite satisfactory for proportional counting. "Geiger" gas (1.3 percent isobutane and 98.7 percent helium) is sometimes used although it is not a common proportional gas. Each of these gases will have characteristically different voltage curves.

The purity of counting gases can affect performance of the counting system. Purity of commercially available counting gases is good, and significant problems are usually not encountered. Each time a new tank of gas is installed the operating characteristics of the counters should be rechecked. If this is not done, plateau shifts and other changes that may result could go undetected.

The most common contaminants in counting gases are moisture, air, and extraneous gases, which can have a serious effect on plateau shapes and other counter characteristics. Air or water vapor is particularly troublesome and will reduce the number of observed counts.

It is desirable and recommended that the gas pressure in the tank of counting gas not be too low. Most laboratories will install a new tank of counting gas when the tank pressure reaches 200 psi. It is important that a good quality two-stage pressure regulator be used on tanks of counting gas.

Another factor is the use of automatic counting systems. When automatic counting systems are purchased, they should be carefully checked to insure that the counting gas passed over samples being counted is exhausted from the counting chamber to the atmosphere or across previously counted samples, and not exhausted into the chambers holding samples not yet counted. If the counting gas is exhausted from the counting chamber over the uncounted samples, there is a distinct possibility of carryover of particulate matter in the counting gas, thereby contaminating uncounted planchets.

Sample Preparation

The preparation of samples for alpha proportional counting is important. There are many methods currently in use for preparation of various types of samples. The methods used will depend on the type of samples, the accuracy desired, and the preferences of the individual laboratories.

Liquid samples, such as river water, are usually evaporated directly on planchets or in beakers from which the resulting dissolved solids are scraped and placed on counting planchets. Samples that have considerable organic matter present, such as river algae, are usually ashed to remove the organic material and then placed on counting planchets.

Solids should be placed on planchets in such a manner that they will adhere to the planchet and not be subject to flaking off by the purging gas. If flaking occurs, the counting chamber will become contaminated, resulting in erroneous counting data. A few drops of lucite-acetone solution will eliminate flaking tendencies. The necessity of obtaining thin uniform sample deposits on planchets cannot be overemphasized.

When samples are filtered on membrane or other types of filters, it is common practice to place such filters directly on the planchets and count the activity present in the solids. The electrostatic charge buildup, which can occur on the filters, may alter the counting efficiencies by as much as 15 percent. When possible, it is best to ash the filters in order to eliminate this problem.

Any sample counted in an internal proportional counter should be completely dry. Any moisture present in the sample can and does absorb electrons readily and can materially affect the resulting count rate. Moisture may also seriously affect counter performance and characteristics.

A number of laboratories have found it best to keep samples in ovens at 105° C. (unless it is suspected that volatile activity is present) until ready for counting. Samples are taken directly from the oven, placed in the chamber, purged with counting gas while cooling, and counted.

The hygroscopic nature of samples should be carefully considered, particularly when the purchase of automatic counting equipment is anticipated. Most commercially available counting systems that count stacks of planchets automatically make no provision for insuring that moisture does not creep into the samples on the stacked planchets. If such a system is not carefully designed to maintain a moisture-free environment for the planchets, this can defeat the purpose for going to an automatic counting system.

If necessary, all sample preparation steps should be continually and carefully evaluated and modified to insure that the data obtained by the instruments are not subject to variations caused by errors in preparation.

Instrument Rooms

Frequently a counting room will contain a wide variety of radioactivity counting instruments. Certain factors should be considered in the design of rooms that are to be used for counting instruments.

The electrical system should be completely separate from other rooms in the immediate vicinity whenever possible. Serious problems may ensue if this is not done. Wiring for counters, motors, ovens, hot plates, and other heavy-duty equipment should be in separate conduits. If this is not the case, transients from such equipment are likely to cause spurious counting.

There should be adequate voltage regulation in all instrument rooms where proportional counting is being carried out. In addition to voltage regulation, it is sometimes desirable to install filters that filter out line transients from the electrical system of the room. Such transients can cause spurious counts to be recorded.

Adequate temperature and humidity control should be designed into an instrument room. Such control is almost a requisite if long-term stability of the instruments is desired.

Whenever possible, preparation of samples should not be carried out in an instrument room, particularly when samples containing a wide range of activity are being analyzed. It is desirable to avoid as much traffic in and out of the counting room as is possible. Some laboratories prohibit smoking, eating, or drinking in counting rooms.

CONCLUSION

Gross alpha analyses have sometimes been referred to as crude analyses yielding very little useful information. However, such analyses are a valuable tool, and if carefully done can yield relationships that are useful in reducing or eliminating the need for specific radionuclide identification.

In addition to gross alpha counting, specific alpha-emitting isotopes of elements, such as plutonium, polonium, radium, uranium, and thorium can also be counted easily by proportional counting techniques, but chemical separations are required.

BIBLIOGRAPHY

SEABORG, G. T. and J. J. KATZ, Editors. The Actinide Elements, National Nuclear Energy Series IV:14B, Chapter 16, McGraw-Hill Book Company, New York (1954).

AMERICAN PUBLIC HEALTH ASSOCIATION. Standard Methods for the Examination of Water and Wastewater, 12th Edition (1965).

CHOPPIN, G. R. Experimental Nuclear Chemistry. Prentice-Hall, Inc., New Jersey (1961).

GLASSTONE, S. Sourcebook on Atomic Energy, 2d Edition. D. Van Nostrand Company, Inc., New Jersey (1958).

FRIEDLANDER, G. and J. W. KENNEDY. Nuclear and Radiochemistry. John Wiley and Sons, Inc., New York (1955).

OVERMAN, R. T. and H. M. CLARK. Radioisotope Techniques. McGraw-Hill Book Company, New York (1960).

O'KELLEY, G. D. Detection and Measurement of Nuclear Radiation (NAS-NS3105). National Academy of Sciences (1962).

PRICE, W. J. Nuclear Radiation Detection. McGraw-Hill Book Company, New York (1958).

ALPHA-SCINTILLATION COUNTERS (USING ZINC SULFIDE PHOSPHORS)

D. Edward Rushing [2]

When an alpha particle bombards an impure crystal of zinc sulfide, a considerable portion of the kinetic energy is transformed into visible light. This effect was discovered soon after the discovery of radioactivity and was used widely in early scientific work as a means of detecting and counting alpha particles. For many years the principle has been applied to the production of self-luminous paints.

Zinc sulfide requires a small amount of "activator" (usually silver) to enable it to produce scintillations more efficiently. When used in counting procedures, it is also necessary to add a "quencher" (nickel) to shorten the duration of the light flash. Such a phosphor is described as silver-activated and nickel-quenched, and it is very efficient as a detector of radiation (*1*).

For many years scintillation counting was done visually and required a great deal of personal attention and patience. Advances in the design of photomultiplier tubes and of the associated circuitry have made scintillation-counting systems electronically simple and practical.

Alpha-scintillation counters consist basically of the following components:

1. A phosphor near, or in contact with, the alpha-emitting substance to be counted and so arranged that the photomultiplier tube can "see" the scintillations. Light-pipes may be used but are not always convenient or necessary.

2. A photomultiplier tube with a diameter equal to or greater than that of the sample. Occasionally, a light-focusing optical system may be used. If this is the case, the photomultiplier tube may

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be considerably smaller than the sample. The photomultiplier tube is covered by a light-tight housing or other cover (*2*), depending on the intended use.

3. A high-voltage supply to furnish voltage to the photomultiplier tube.
4. An amplifier that picks up the electrical pulses from the photomultiplier tube and amplifies them or acts as an impedance changer.
5. A discriminator or gain setting that rejects pulses smaller than a predetermined size.
6. A scaler that counts the pulses that pass the discriminator.
7. Optional features, such as preset time and preset count.

Mounting and Counting Techniques

Features of Scintillation Counting of Alpha Particles

Some features of scintillation counting of alpha particles are:

1. Samples of up to 5 inches in diameter may be counted with suitable equipment.
2. Scintillation counters are simple compared to proportional counters and the older types of radon 222 counters.
3. A trace of water in the sample ordinarily does not interfere with the counting process.
4. Backgrounds are about equal to those obtained, in practice, with proportional counters.
5. Some types of detectors (for example, radon 222 scintillation cells) are readily removable from the counting system. This permits measurement of high or low activities without danger of cross-contamination.
6. The cost of the counting system is comparable to that of an internal proportional counting system.
7. Counting gases are not required in routine work.
8. The efficiency of counting varies with the physical arrangements and the application of the phosphor. When the planchet is near the phosphor, the efficiency is 36 to 40 percent (*3*). When the phosphor and sample are arranged so that both sides can be "seen" by photomultiplier tubes, the efficiency is about 90 percent (*4*), but this arrangement requires a more expensive counter. In the case of radon 222 counting, the efficiency is about 80 percent (*5-6*).

9. The detector must be protected from light by a light-tight housing. In portable equipment, a thin opaque material is used (*2*). Some counters are arranged so that samples can be changed without exposing the photomultiplier tube to light, making it unneccessary to turn off the high voltage during the operation.

Special Requirements for Sample Preparation

Sample preparation is about the same as for alpha counting in proportional counters. Self-absorption by the sample (and in some arrangements by the phosphor) is as important a consideration as it is with a proportional counter.

Counting of Gases (Radon 222)

Alpha-scintillation counting is applied to counting in both the gaseous and solid phases. In the counting of gases (radon 222), the sample container may be a bottle or flask coated on the inside with the phosphor. In semiquantitative work the entire inner surface may be coated (*7-9*). For more refined work, the bottom of the vessel should be flat and transparent since this type of vessel transmits larger and more uniform pulses of light (*5, 6, 10*). The following facts should be kept in mind in the design and use of such counting cells:

1. Absorption of alpha particles by the gaseous medium should be kept at a minimum consistent with practical use of the cells. Such absorption can be reduced, in theory, by: (a) minimizing the dimensions of the cell holding the volume constant and (b) using a low-density gaseous medium. Procedure (a) would lead to a near-spherical design that does not appear sound from the optical and practical points of view. Procedure (b) indicates the use of helium (hydrogen is too dangerous) as the gaseous medium for the use of reduced pressures. Helium is practical and gives the best results, although radon 222-free air or nitrogen in a small cell is practical and is much easier to use. Operating at reduced pressures is inconvenient and reduces the amount of gas available for deemanation of a radium 226 bubbler (which is one of the chief uses for the technique). In effect, it would be just as well to use a smaller counting cell and operate at atmospheric pressure.

2. Degradation of the alpha energies by any binder used should be minimized. Nonuse of a binder is possible, but the cells have to be evacuated and handled carefully to avoid dislodging the phosphor.

3. In order to secure the maximum pulse size, it is necessary that the phosphor coating be of definite minimum thickness (about 20 mg/cm^2) (*5*).

4. Charges may build up on glass and silica parts of the counting vessel, especially if the cathode of the photomultiplier tube is not grounded in the electronic circuitry. This is remedied by using a metal container with a conductive silica window (coated with tin oxide); the entire assembly is grounded to the grounded chassis. Charges on the window vary and affect the behavior of the charged radioactive daughters in the cell, thereby giving erratic counting efficiencies (*5*).

5. Backgrounds of cells are important unless they are insignificant in comparison to the activities being counted. They are affected by the materials of construction, discriminator settings, contamination by previous samples, and indirectly by the gaseous medium used. Contamination by previous samples is roughly proportional to the product of the activity and time the sample is in the cell. In the case of radon 222, the long-term increase in background is caused by the growth of polonium 210 from lead 210, and it grows slowly over a period of weeks or months. Other decay products of radon 222 deposit in the cell, but die out with an approximate half-life of 30 minutes once the parent (radon 222) is removed. Cells should be flushed thoroughtly after use.

The alpha backgrounds can be divided into five components:

1. Alpha emitters present in materials of construction.
2. Radon 222 remaining from previous sample (adsorbed and/or dissolved).
3. Short-lived decay products of radon 222.
4. Long-lived decay products of radon 222.
5. Radon 222 present in counting gas (from radium 226 in gas cylinder, valve, and connecting tubing).

Since radon 222 is soluble in many organic media, organic substances within the cell (binder, grease, and so forth) should be reduced to a minimum. Rubber will pick up radon 222 to some extent, but with a short time of contact the loss is not noticeable on the sample being analyzed. However, if the sample is high relative to the succeeding sample, the next sample count may be affected, particularly if it is low.

Counting of Solid Preparations

When solid samples are counted by the scintillation technique, several physical arrangements may be used:

1. The solid sample is deposited on a planchet (as for a proportional counter), and the planchet is placed close to a phosphor-coated screen (phosphor on the adjacent side of the glass). The screen is optically coupled to the photomultiplier tube; in fact, the face of the photomultiplier tube is sometimes coated with the phosphor and serves as the screen. The photomultiplier tube is mounted vertically, face down, in a light-tight housing. A sliding-drawer arrangement can be used to avoid exposing the photomultiplier tube to light. The phosphor screen normally can be reused. The use of helium or an evacuation technique will reduce absorption of alpha particles in the gas-filled space between the sample and the detecting phosphor-coated screen, but ordinarily neither of these techniques is used.

The sample precipitate on a filter may be mounted on a ring-and-disc arrangement in intimate contact with the phosphor-coated sheet of plastic (*3*). By using phosphor-coated plastic on both sides of the sample, better counting efficiency can be obtained (*4*).

Certain alpha emitters may be deposited electrochemically on metal discs. These may be counted by either of the techniques just mentioned.

2. The solid sample in the form of a precipitate (for example, radium 226-barium sulfate) is intimately mixed with the phosphor and reproducibly spread on a screen or placed in a glass vessel (glass planchet or Microslide® with depression). In the latter case, the photomultiplier tube should be mounted vertically with its face up. In a variation of the technique, the sample (if essentially weightless) can be evaporated in the glass planchet and the phosphor can be placed on top of the sample.

3. In exceptional cases, it is possible to deposit the desired alpha emitter on the phosphor by stirring the phosphor into a solution of the substance. The radionuclide is deposited chemically, is separated by centrifuging or filtering, and then is counted as in 2 above. An example of this is the deposition of polonium 210 from solution on to the ZnS phosphor.

Electronic Equipment

Scintillation-counting equipment for alpha detection is relatively simple. The pulses are large enough so that little amplification is needed. Some problems such as electronic noise do arise.

A diagram of a typical alpha-scintillation counter is shown in figure 4.

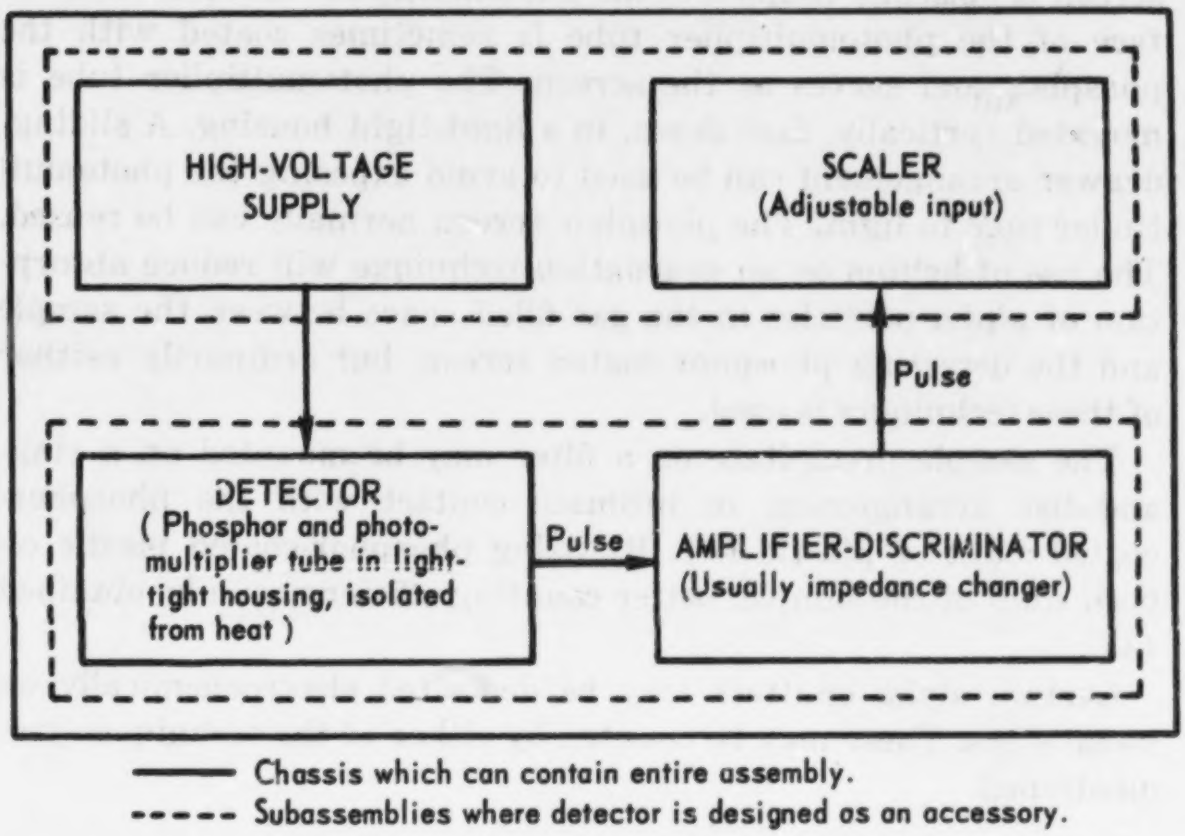
HIGH-VOLTAGE SUPPLY
SCALER (Adjustable input)
Pulse
DETECTOR (Phosphor and photo-multiplier tube in light-tight housing, isolated from heat)
Pulse
AMPLIFIER-DISCRIMINATOR (Usually impedance changer)
Chassis which can contain entire assembly.
Subassemblies where detector is designed as an accessory.

Figure 4. Diagram of a scintillation counter for alpha particles.

High-Voltage Supply

It is customary to supply the high voltage to the anode (or cathode) and to the dynodes of the photomultiplier tube by means of a series of voltage-dropping resistors. This means that a steady current flows in the high-voltage system. The counting system must not produce spurious pulses under these conditions. The operating and permissible voltages depend on the photomultiplier tubes used. A variable voltage range should be provided to permit adjustment for a maximum signal-to-noise ratio.

Amplifier-Discriminator

With vacuum tube circuits, the amplifier-discriminator usually consists of a cathode-follower and a discriminator circuit similar to those used in proportional counters. Transistor circuits serving the same purposes can be used.

Scaler

Two decades of scaling are quite satisfactory for most samples likely to be encountered in surveillance and are satisfactory for checks with pulse generators and standards of reasonable activity. An automatic cutoff for the preset count may be incorporated. An odometer type of clock is useful, and the minimum reading should be 0.1 or 0.01 minutes depending on whether relatively long or short counting periods are to be used. For long counting periods (overnight), a built-in clock is convenient but not necessary if the counting is to be stopped manually.

Photomultiplier Tubes

Flat-faced photomultiplier tubes generally are used in scintillation counting. For most alpha counting applications, 2- or 3-inch photomultiplier tubes are satisfactory. Photomultiplier tubes may have, or develop during use, sufficient noise to give spurious counts. Specifications provided by manufacturers do not provide sufficient information to serve as a basis for rejection of such tubes. Noise pulses can be discriminated against because the alpha pulses are much larger, and therefore the selection of tubes is not important here as it is in beta- and gamma-scintillation counting. The noise level of a photomultiplier tube rises with increase in temperature, but again this factor is not important in alpha counting. The design of the instrument should place the photomultiplier tube in a position where it will operate at room temperature or no more than a few degrees above room temperature.

The photomultiplier tube should not be exposed to light immediately (an interval of 3 to 4 hours should be used as a norm) before use even if the high voltage is off, and it must never be uncovered with the high voltage applied. A high-voltage safety switch should be opened automatically when the light cover is removed to avoid damage to the photomultiplier tube.

Scintillation-Counting Accessories for Proportional Counters and Scalers

Scalers or counters can be used with a variety of scintillation-counting accessories that are designed for specific purposes (for example, radon 222 measurement). Such accessories for counting solid samples are commercially available and can be custom made for radon 222 counting. These accessories should not cost more than a few hundred dollars. At very low levels of radioactivity, electronic noise may be a problem with certain instruments because of unsuitable high-voltage supplies.

General Specifications

The general specifications of the electronic equipment have to be considered in connection with the intended use of the equipment. The manufacturer cannot be held responsible for the overall performance of the counter-detector system unless he supplies the detectors, phosphors, and planchets. The system specifications for such a counter are centered around two main items: (1) the background, which is made up of electronic noise (spurious counts), and a component resulting from interactions between alpha-emitting substances and the phosphor, and (2) the length and slope of the plateau that are obtainable with the particular detector. The electrical characteristics of the system are intimately related to the nonelectric features of the detector. Some of these relationships are difficult to specify without recourse to experiment or to work done by other persons. It is possible to specify the electronic noise of a counter at various voltages, but unless the satisfactory operating voltage, as determined by the plateau, is known, such noise specifications may be more severe than the application demands. The length, position, and slope of the plateau are determined to a greater or lesser degree by the type of photomultiplier tube, the physical characteristics of the detector, and the design and adjustments of the counter. In the determination of radon 222, the optimum operating voltage may vary as much as 150 volts with different photomultiplier tubes of the same type. The electronic noise increases rather rapidly with increased operating voltage. With careful design and construction, the electronic noise may be as low as 1 count per 16 hours; however, electronic noise of 2 counts per hour at the operating voltage is satisfactory for low-level work.

A carefully designed and adjusted scintillation-counter assembly for counting alpha particles (radon 222) will have a plateau similar to that of a proportional counter. The length and slope will be comparable to those of a proportional counter (table 1).

TABLE 1. *Radon count rate as a function of voltage applied to photomultiplier tube*[a]

Volts	Corrected[b] counts ($\pm\sigma$)[c] per 10 minutes	Percent ($\pm\sigma$) (1,250 volts = 100)
850	4,130± 65	47.3±0.7
900	6,721± 82	77.0±0.9
950	7,843± 89	89.8±1.0
1,000	8,250± 91	94.5±1.0
1,050	8,527± 93	97.7±1.1
1,100	8,773± 94	100.5±1.1
1,150	8,749± 94	100.2±1.1
1,200	8,704± 94	99.7±1.1
1,250	8,732± 94	100.0±1.1
1,300	8,730± 94	100.0±1.1
1,350	8,736± 94	100.0±1.1
1,400	8,616± 93	98.7±1.1
1,450	8,845± 94	101.3±1.1
1,500	12,407±112	142.1±1.3

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[a] Dumont 6363 tube. Scaler sensitivity set to accept 0.15 volt.

[b] Corrected for decay of radon.

[c] σ is the standard deviation.

Scintillation cells for radon 222 counting are available with a background of 5 counts per hour when used with helium. The background may be doubled or tripled if aged air is used in the cells. The improved backgrounds with helium are possible because a larger net pulse is obtained when helium is substituted for air in the scintillation cell. This fact makes it possible to set the discriminator at a higher level; thus eliminating more of the noise and very low-energy alpha pulses whose points of origin are probably within a phosphor itself.

Maintenance

Maintenance is usually limited to checking and replacing tubes and to adjusting sensitivity. These operations can be performed by personnel familiar with other types of counters. The accuracy of counters should be checked periodically by counting standards. Plateaus should be determined occasionally. Maintenance is not a serious problem with scintillation counters.

Special Requirements for Wiring and Housing

Space and power requirements for scintillation counters are essentially the same as for proportional counters. They should be protected from radioactive contamination, chemicals, fumes, heat, and high humidity. A constant-temperature room is desirable.

Availability

Although some alpha-scintillation counters are commercially available, most are custom made. Beta-scintillation counters are commercially available and many can be used for alpha counting with major adjustments (*4*). These systems are more complicated and expensive than need be for simple alpha counting.

Cost of Equipment and Accessories

Scaler attachments for scintillation counting of solids are commercially available. Attachments for scintillation counting of radon 222 can be custom made. The cost of these items is a few hundred dollars. The scintillation cell required for radon 222 counting must be purchased as a separate item.

A complete scintillation counter (for radon 222) with a 3-inch photomultiplier tube and two decade units can be purchased (custom made) for about $1,100.

Scintillation cells for radon 222 are usually made in the laboratory. The parts for these cells cost about $15 per cell. These cells are available commercially for about $200. Bubblers for radon 222 standards and radium 226 samples cost about $22 each.

REFERENCES

(*1*) KALLMAN, H., M. FURST, and M. SIDRAN. Scintillation counting techniques. Nucleonics 10:15 (1952).

(*2*) GRAVES, J. D., L. A. WEBB, and R. H. DAVIS. Performance of lucite-bonded alpha-scintillation screens. Nucleonics 10:68 (1952).

(*3*) HALLDEN, N. A. and J. H. HARLEY. An improved alpha-counting technique. Anal Chem 32:1861 (1960).

(*4*) SAX, N. I. and D. ROSI. 4-pi zinc sulfide alpha counting. Anal Chem 32:1900 (1960).

(*5*) LUCAS, H. F. Improved low-level alpha-scintillation counter for radon. Rev Sci Instrum 28:680 (1957).

(*6*) RUSHING, D. E., W. J. GARCIA, and D. A. CLARK. The analysis of effluents and environmental samples from uranium mills and of biological samples for radium, polonium, and uranium. Radiological Health and Safety in Mining and Milling of Nuclear Materials II:187 (1964). International Atomic Energy Agency, Vienna.

(*7*) KUSNETZ, H. L. Radon daughters in mine atmospheres: a field method for determining concentrations. Amer Industr Hyg Ass J 17:85 (1956).

(*8*) VAN DILLA, M. A. and D. H. TAYSUM. Scintillation counter for assay of radon gas. Nucleonics 13:68 (1955).

(*9*) HOLADAY, D. A. et al. Control of radon and daughters in uranium mines and calculations on biologic effects. PHS Publication No. 494. Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402 (1957).

Chapter II. Beta-Counting Systems

GEIGER-MUELLER COUNTERS

Richard K. Stoms [1]

A large variety of instrument types are available for the measurement of nuclear radiation. The choice of a type of system or a specific instrument for a specific application is generally not too difficult. Difficulty does arise, however, when a single instrument type must be selected to perform several different measurement jobs. In making a selection, the Geiger-Mueller (GM) counter is often considered.

Although it is beyond the scope of this chapter to present a detailed discussion of the theory involved in Geiger-Mueller counters, a brief review on the operating principles follows.

The Geiger-Mueller counter detects nuclear radiation by gas ionization. It consists basically of an envelope containing two electrodes and a filling gas. The cathode is usually part of the envelope, the anode is usually a wire or wire loop from one- to five-thousandths of an inch in diameter, mounted centrally within the cathode. Noble gases such as argon, helium, or neon are commonly used, and generally a small percentage of some other gas is added to provide quenching.

In a Geiger-Mueller counter, an ion pair produced in the gas by incident radiation will initiate the discharge. The amount of energy required is quite small. One or more ion pairs may be produced in the gas as a result of direct energy absorption by the gas or as a result of secondary electrons emitted from either the anode or cathode. The electrons produced will move toward the anode. Since the anode has a small diameter, the electric field will have a high gradient in its vicinity. The field gradient will accelerate the electrons which will ionize additional gas molecules because of their velocity. Electrons from these ions will in turn cause more ionization. Most of this electron multiplication will occur in the vicinity of the anode where the electric field gradient is highest. When the number of electrons produced and collected in this manner is independent of the number of ion pairs formed in the initiating event, the tube is operating in the Geiger-Mueller region.

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As electrons are collected, positive ions will be left in the vicinity of the anode. When the positive ion density is great enough to reduce the electric field below the value required to produce avalanching, the discharge will terminate. (Avalanching is the building up of ionization by the process of gas multiplication to the point of a momentary discharge.) The production of the discharge—until it is terminated by space charge—requires a fraction of a microsecond.

The positive ions in the space charge around the anode will then move relatively slowly to the cathode. As the ions are neutralized at the cathode, electrons may be produced in the gas and cause additional discharge. This results in a continuous discharge. In order for the tube to be useful, the discharge must be terminated after an ionizing event. This is done externally in the electronic circuitry by reducing the voltage on the tube after a discharge has occurred or by introducing a quenching agent in the gas. Virtually all Geiger-Mueller counters at present use internal quenching. (Quenching may de defined simply as the prevention of an undesirable secondary discharge caused by migration of positive ions to the cathode.) Either organic or halogen quenching gases may be used. Ethyl alcohol and ethyl formate are two organic gases employed, while bromine and chlorine may be the halogens used.

The quenching gas used must have a lower ionizing potential than the gas used as the main component of the mixture. If this is the case, the positive ions, as they migrate to the cathode, will transfer their charge to the quenching gas. Only quenching gas ions will reach the cathode and be neutralized. The energy released when this occurs will dissociate the quenching gas molecule rather than produce further ionization. The molecular fragments of the organic gases apparently do not recombine, and the quenching gas is depleted. In general, organic-quenched tubes have a useful lifetime of about 10^{10} counts or less. Recombination of the fragments of the halogen molecules prevents depletion in this way, and much longer lifetimes may be expected when halogens are used. For this reason halogen-quenched tubes may be operated at higher relative voltages without serious decrease in lifetime. The result is a larger output pulse from the halogen-quenched tube (approximately 10 volts versus approximately 1 volt for the organic-quenched types).

A Geiger-Mueller tube is rendered insensitive to incident radiation during the initial stages of a pulse. This period, during which another pulse cannot be generated, is called the "dead time." Positive ions are present for a short time immediately following the dead time. They act to reduce the electric field between the electrodes, and until neutralized they reduce the amplitude of pulses generated. This is defined as the "recovery time." The dead time and recovery time vary even with the same tube, and the position of the line that separates them depends on the associated circuitry. For these reasons, the circuitry is adjusted to give a constant inoperative interval that is a little greater than the dead time of the Geiger-Mueller tube. The inoperative interval of an instrument is usually about 10^{-4} seconds.

Geiger-Mueller counters are inexpensive. Associated electronic circuitry is less complex because of the large and reasonably uniform pulses. The count rate, limited by positive ion collection, is within the capabilities of very simple rate meters and scalers. This, coupled with the very low threshold of sensitivity, provides the basis for which Geiger-Mueller counters are selected for a specific application.

With improvements in electronic circuitry and components, the advantages of Geiger-Mueller counters over other types of counters have been reduced. The apparent advantage of a uniform output pulse from the detector is nullified if there is a requirement to differentiate between the energies of two or more types of primary incident particles or quanta.

The window-type Geiger-Mueller counter is quite useful as a beta counter and is specifically recommended when low-activity beta samples (except extremely low-energy radionuclides such as carbon 14 and tritium) are to be counted. The counter for this application should provide for samples in 2-inch-diameter planchets. For low backgrounds, the detector will be shielded with either lead or steel and have an "umbrella" (usually Geiger-Mueller tubes) operated in anticoincidence. Backgrounds are normally between 0.5 and 1.0 counts per minute for a chamber with a 2-inch diameter. The detector is invariably a gas-flow type, and a window is recommended to minimize contamination of the sensor. Efficiencies of the order of 40 percent for strontium 90 are realized with this configuration.

One of the problems in specifying or evaluating Geiger-Mueller counters is in stating exactly how the efficiency is to be determined. The efficiency for a point source at the center and very close to the window will be substantially higher than that obtained with the source distributed over the bottom of a 2-inch-diameter planchet and five-sixteenths of an inch below the window. Furthermore, the efficiency will fall off as a point source is moved radially from a position below the center of the window. If the efficiency drops off too rapidly, errors can be introduced because of nonuniform distribution of the source or sample material in a sample holder. The count rate in a good instrument will not drop more than 5 percent when a point source is moved three-fourths of an inch from the center of a chamber having a diameter of 2 inches. Some chambers having a decrease of 20 percent appear to be adequate although one must realize that the error involving sample distribution in the sample holder will be higher.

Window thickness is a factor influencing efficiency. A lower density window will attenuate beta particles less and result in a larger number entering the sensitive volume with sufficient energy to be counted. Extremely thin windows will increase the probability of accidental rupture and dimensional variations because of pressure changes that may influence chamber geometry. No specific recommendation can be made as to the best density to use since this will depend to a large extent on the threshold of sensitivity of the chamber, the dead zone adjacent to the window, and the energy of the beta particles to be counted. A density of 0.9 mg/cm^2 is typical for counting strontium 90 and yttrium 90.

A proportional counter fitted with a window, shielding, and an anticoincidence "umbrella" will provide as low a background and a very slight increase in efficiency over the Geiger-Mueller counter. The slight increase in efficiency hardly warrants the additional cost of the proportional counter. Admittedly, the proportional counter will be able to count alpha particles in the presence of beta particles, but this can be performed easily on an unshielded internal proportional counter at higher efficiency.

The discussions of sample requirements and counting efficiency given in this chapter under "Proportional Counters" are applicable to Geiger-Mueller counters which use window-type detectors.

CONCLUSION

Geiger-Mueller counters are available in many forms from simple survey meter types to precision laboratory instruments. The types in which the gas is permanently incorporated in the tube are not useful in low-level applications where low count rates are obtained and backgrounds must be low. An exception is their application in a guard or "umbrella" around another counter.

A gas-flow Geiger-Mueller counter with a window, shielding, and anticoincidence guard or "umbrella" is used for counting of low-activity beta-emitting samples. It is preferred to the low-background proportional counter for this application on the basis of virtually equal performance and lower cost and complexity. The low-background proportional counter will be more desirable if alpha particles are to be counted in the presence of beta particles and an internal proportional counter or ZnS (Ag) counter is not available.

BIBLIOGRAPHY

PRICE, W. J. Nuclear Radiation Detection, Chapter 5. McGraw-Hill Book Company, New York (1958).

WILKINSON, D. H. Ionization Chambers and Counters, Chapter 7. Cambridge University Press, London, England (1950).

ZUMWALT, L. R. U.S. Atomic Energy Commission Document, AECU-567. U.S. Atomic Energy Commission, Washington, D.C. (1950).

SARPE, J. Nuclear Radiation Detectors, Chapter 6. John Wiley and Sons, Inc., New York (1955).

SNELL, H. Nuclear Instruments and Their Uses, Chapter 5B. John Wiley and Sons, Inc., New York (1962).

WATT, D. E. and D. RAMSDEN. High Sensitivity Counting Techniques, Chapters 2.8 and 8.5. The MacMillan Company, New York (1964).

PROPORTIONAL COUNTERS

Charles R. Phillips [2]

The proportional counter is used primarily for detecting alpha and beta radioactivity. The ability to detect both alpha and beta particles and to distinguish between the two makes the proportional counter particularly attractive to those laboratories operating on a limited budget.

The Geiger-Mueller (GM) and proportional counters are the most widely used instruments for beta counting. The proportional counter is capable of higher count rates than the Geiger-Mueller counter. This is normally of little consequence in the counting of environmental samples because of the low activities usually associated with such samples.

Proportional counters may have either thin windows through which radiation is admitted or no windows. In the windowless-type counter the sample is placed inside the counting chamber, and they are usually referred to as internal proportional counters. Internal proportional counters have a greater efficiency than window-type counters because of improved geometry and the absence of air or window between the sample and active volume of the detector to degrade the particles' energy. Windowless-type counters are easily contaminated. The sample must be in electrical contact with the counting chamber walls, and they are more time-consuming to operate since the counting gas must be purged through the chamber to eliminate any air that may have entered when the sample was placed in the chamber. The advantages and disadvantages of window-type counters versus windowless-type counters must be weighed according to the energy of the beta particles to be detected. With low-energy particles, such as carbon 14, a windowless-type counter is particularly recommended.

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The proportional counter is operated in the proportional region of the "pulse-height versus applied-voltage curve" (ch. I fig. 2). In this region gas-amplification is employed while pulse-height proportionality to the amount of primary ionization produced by the radiation is retained.

Proportional counters usually employ "P-10" gas—a mixture containing 90 percent argon and 10 percent methane. They are usually of the "gas-flow" type; that is, gas is continuously flowing through the counting chamber. This method maintains a positive pressure in the detector and aids in preventing the entrance of dust, air, and so forth, into the detector.

Detectors that will accommodate 2-inch-diameter sample planchets are recommended for counting activity in the 1 to 10,000 pCi range.

Proportional counters are usually purchased as complete systems consisting of: (1) a shielded detector, (2) a preamplifier, (3) a pulse discriminator, (4) an amplifier, (5) a scaler, (6) a high-voltage supply, (7) a timer, and (8) a gas-flow system consisting of pressure regulator, flow-rate control valve, and flow-indicating bubbler.

Performance Specifications

Background

The background counting rate in the beta plateau comes primarily from cosmic and gamma radiation (*1*). The background count rate of a given counter is a function of the amount of shielding, cosmic-ray flux, contamination in the chamber construction materials, detector size, level of radiation in the immediate area, and whether the counter has an anticoincidence background reduction system.

A substantial reduction in background can be achieved by anticoincidence counting techniques. This is accomplished by placing a second detector system (called a guard detector) around the proportional detector. A count from the detector will not be recorded if a simultaneous count is produced in the guard detector.

Beta counters equipped with anticoincidence systems and massive shielding are referred to as "low-background beta counters." Low-background beta counters are presently available with backgrounds of less than 1 count per minute. Proportional counters without anticoincidence background reduction and shielding exhibit beta backgrounds of the order of 50 counts per minute with a 2-inch diameter detector.

Counting Efficiency

Counting efficiency is a function of geometry, backscatter, window absorption, and self-absorption. The efficiency is also dependent on the thickness of the detector window. If these factors are held constant, beta-counting efficiencies are a function of beta-particle energies.

An important consideration in counting samples other than point sources (most environmental samples are not point sources) is that the detector exhibits uniform efficiency over the entire area to be subtended by the sample.

The counter should be efficient enough for each beta energy to be counted in order to eliminate unduly long counting times and to achieve the desired statistical accuracy.

Plateaus

Beta plateaus of a proportional counter are not as flat as alpha plateaus because of the inherently large distribution in the energy of beta particles (*1*).

Plateaus may be described in two ways: (1) their lengths in volts and (2) their slopes. It is desirable to have plateaus of at least 200 volts in length with a slope of not greater than 2 percent per 100 volts.

General

Additional features desirable in a proportional beta counter are:

1. A system should have an adjustable preset timer capable of presetting times up to 500 minutes.

2. The system's performance should not be adversely affected by normal line voltage variations.

3. Each scaler should be capable of accepting total counts of at least 10^5 and counting rates of 10^4 to 10^5 counts per second with a double-pulse resolution of the order of 10 microseconds.

4. The detectors should be easily removable for decontamination.

Operational Aspects

Sample Requirements

The sample geometry, planchet material, and absorbing medium should be consistent with those for which the detector was standardized. Uniform deposition of the sample over the entire area should be attained. To minimize contamination of the counter, the sample should contain no loose solids.

REFERENCE AND BIBLIOGRAPHY

(*1*) PRICE, W. J. Nuclear Radiation Detection. McGraw-Hill Book Company, New York (1958).

SNELL, A. H., Editor. Nuclear Instruments and Their Uses, Volume I. John Wiley and Sons, Inc., New York (1962).

SEGRE, E., Editor. Experimental Nuclear Physics, Volume I. John Wiley and Sons, Inc., New York (1953).

GLASSTONE, S. Sourcebook on Atomic Energy, 2d Edition. D. Van Nostrand Company, Inc., New Jersey (1958).

BETA-SCINTILLATION COUNTERS

Joseph C. Drobinski, Jr.[3]

Liquid-scintillation and plastic-scintillation counting of beta particles are covered in this section. Their detection by organic crystals, inorganic crystals, glass, and static-gas scintillation devices has purposely been omitted as they are beyond the scope and intent of this report.

Liquid-Scintillation Counting

As mentioned previously, the detection of beta particles usually is accomplished by using Geiger-Mueller and proportional-type counters; however, all the beta-emitting radionuclides, such as zirconium 95, niobium 95, iron 59, samarium 151, rubidium 87, and calcium 45, can be readily counted by liquid-scintillation techniques. In particular, samples of weak beta emitters, such as carbon 14, tritium, plutonium 241, nickel 63, and rubidium 106, can be best detected by liquid-scintillation or gas-phase counting.

Aromatic organic solvents, such as benzene, xylene, toluene, and phenylcyclohexane, are used as the basic solvents. Dioxane, a cyclic ether, is a better solvent with polar compounds (for example, water or aqueous suspensions of solids). Chemical compounds of the highest purity available (scintillation grade) should always be used to avoid quenching problems.

Pure organic crystalline compounds soluble in the solvents mentioned above are used as fluors or phosphors that emit short-lived flashes of light (scintillations) when the beta particles of the radionuclide-containing compounds interact with them. There are dozens of these "scintillators" (usually of an aromatic type), as well as the auxiliary ones called "secondary solutes," that displace the fluorescence toward longer wavelengths in the visible blue region where many light-detecting photomultiplier tubes have the highest spectral sensitivity.

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It is preferable to have matched photomultiplier tubes in electronic coincidence arranged so it is possible to discriminate against Compton recoil electrons and cosmic particles, background, and/or another radionuclide in the sample. The most sensitive tube can be placed to monitor the sample and the pulses of the second tube can be transmitted to scaling circuits to record light that both photomultiplier tubes "see." In this manner, the greatest sensitivity,[4] reproducibility, and the highest figure of merit can be achieved.

Although much of the scientific literature is concerned with liquid-scintillation counting of carbon 14 and tritium, any beta-emmitting radionuclide can be counted in a system if it can be made a part of, be suspended in, or externally excite the scintillation medium (*1*). Horrocks (*1*) has grouped the means of incorporating radionuclides as follows:

1. Organic complexing agents.
2. Salts of organic acids.
3. Dissolution of aqueous solutions in alcohols, dioxane base mixtures, and naphthalene solutions.

Ronzio (*2*) studied the scintillation efficiency of various scintillating solutions containing several metal salts of naphthalene and 2-ethyl hexanoic acid, which are soluble in aromatic hydrocarbons. The octoates of cadmium, lead, bismuth, and uranium were prepared by Ronzio. Rubidium 87, calcium 45, samarium 147, potassium 40, and sodium 22 can be assayed by these techniques.

Aqueous solutions have been counted by using a ternary or higher mixture after the aqueous solution is mixed with alcohol. Benzene or toluene could be the third component. Dioxane is frequently used since it is miscible with water. Since even small amounts of water and oxygen or oxygen-containing compounds "quench" the light output of the scintillator, care must be exercised and judgment used as to the amounts to be added and the subsequent degree of quenching to the light, which in turn affects the minimum detectable activity.[5]

[4, 5] The International Commission on Radiological Units and Measurements defines this as the minimum detectable activity (MDA). Radioactivity: Recommendations of the International Commission on Radiological Units and Measurements (1962), ICRU Report 10 c, National Bureau of Standards Handbook 86, p. 26. Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402 (1963).

Finely divided solids suspended by gelling agents in liquid-scintillation solution have surprisingly high-counting efficiencies. Powdered Thixcin (*3*) and SiO_2 (Cab-O-Sil) (*4*) have been used successfully. Cerenkov radiation in aqueous solution produced by high-energy beta emitters can also be observed with a radionuclide like magnesium 56 (2.86 MeV) (*5*).

The analyst and investigator have a valuable and versatile tool with liquid-scintillation counting. It is possible to count samples routinely and quickly without the problems of precipitate thickness, uniformity of a counting planchet, cross-contamination, and extensive radiochemical separation. The method is very flexible and can accommodate a wide activity range. Each worker can devise, according to the need, his own counting mixture by mixing solutions, phosphors, and samples. It is useful for the radiochemist, the biomedical research worker, and those interested in environmental levels of radioactivity. The liquid-scintillation method has been used successfully by Arnold (*6*) and Pringle (*7*) to establish radiocarbon dates.

The use of the figure-of-merit expression, either S^2/B or E^2/B, will aid the user of the technique and instrumentation. The background rates, B, vary with temperature, the materials of construction of the sample vial and the thermal noise of the photomultiplier tubes. S is the sample counting rate and E is the counting efficiency.

A common procedure among instrument manufacturers has been to cool the sample and detectors in a freezer (−10° C. to 5° C.), although several firms have ambient temperature instruments. Thermoelectric cooling has been proposed for another instrument. In environmental radioassay, it is preferable also to use quartz, polyethylene, or low-potassium glass sample vials. Optical coupling (*8*) of the photomultiplier tubes and counting vial with silicone grease, light pipes, and reflective surfaces improve the overall efficiency, E.

Common sample vial sizes are 20, 40, and 60 milliliters. Hours and Kaufman (*9*), for their low-level tritium work (500 tritium units),[6] used special 180-milliliter vials. It is preferable to increase the counting rate, S, if other factors are constant or minimal.

It is also useful to have a yardstick with respect to number of samples to be assayed and intervals of counting time (*10*). Several instruments automatically reject samples below a selected counting rate. The recent innovation of pulse summation techniques to eliminate preamplifiers on the photomultiplier tubes has lowered backgrounds and improved detection sensitivity considerably.

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[6] One tritium unit = 1 atom of $^3H/10^{18}$ atoms of H.

Excellent reviews with extensive references by Davidson and Feigelson in 1957 (*11*) and Rapkin in 1963 (*12*) exist to orient the beginner and guide the established workers in the field.

Completely automatic liquid-scintillation spectrometry systems are commercially available for about $13,000. Sample capacity varies from 40 to 400 sample vials. Manual and semiautomatic instruments can be purchased from $4,000 to $8,000. The majority of instruments made in the United States are transistorized. Typical background rates for carbon 14 are 12 to 20 counts per minute. The background for a weak beta emitter, such as tritium, is in the 25 to 45 counts per minute range since the voltage on the photomultiplier tube normally is higher than for carbon 14.

Efficiencies increase with increasing energy. A Packard Tri-Carb 314X, for example, would count with efficiencies in the aqueous phase of 8 to 10 percent for tritium, 15 to 20 percent for nickel 63, and 30 to 40 percent for carbon 14. In an organic mixture the efficiencies could be 40 percent, 70 percent, and 90 percent, respectively, for the same three radionuclides. The problems of self-absorption, windows, geometry, and reproducibility of sample preparation are eliminated or greatly reduced if this technique is used.

Plastic Detectors

In 1950 Shorr and Torney (*13*) discovered that scintillation occurred in solid solutions of many substances embedded in polystyrene (*14, 15*). The search for better solid solutions has continued. Plastic scintillators have distinct and considerable advantages over liquids, flow counters, and organic crystals. Some of these advantages are:

1. Ease of sample preparation.
2. Good optical qualities, mechanical strength, and thermal qualities (some may be used in the range of −196° C. to +70° C.).
3. Since in some cases plastics can be highly polished, reflectors can be omitted.
4. It is possible to introduce wavelength shifters, as in liquid-scintillation counting, to match more closely the spectral response of the photomultiplier tube.
5. Some plastics can be used in a vacuum, which is not the case with most organic crystals.

6. Although the light yield is less, the fluorescence time is short, permitting the use of plastic scintillators in fast coincidence circuits in the nanosecond region.

7. No protective coating is necessary in some cases.

8. They are machinable to almost any reasonable geometry.

Spiral flow cells and spheres are commercially available (*16*). Polystyrene and polyvinyl toluene are two of the best plastics to use. In liquid-scintillation counting, the best activators for high efficiency are the same organic heterocyclic compounds with three or more aromatic rings combined in condensed or linear configurations (table 1) (*14*).

TABLE 1. *Scintillation efficiencies of some plastic scintillators* [a]

Activator and shifter of spectrum (in polystyrene)	SE relative to stilbene (percent)
TP (2 percent) +α NPO (0.1 percent)	82
PPO (2 percent) +α NPO (0.1 percent)	77
TP (2 percent+POPOP (0.2 percent)	86
PPO (2 percent) +POPOP (0.2 percent)	79
TP (1 percent) +PPO (1 percent)	81
TP (2 percent) +BBO (0.1 percent)	86
POPOP (1 percent)	62
α NPO (1 percent)	63
BPO (0.5 percent)	70
PB (1 percent)	71
BBO (0.5 percent)	65
PPO (1 percent	70
TP (2 percent) +triphenylpyrazoline (0.04 percent)	85
TP (3 percent) +TPB (0.02 percent)	78

[a] Reference (*14*).

TP: p-terphenyl
BBO: 2,5-di(4'-biphenylyl) oxazole
BPO: 2-(4'biphenylyl)-5-phenyloxazole
NPO: α-naphthylphenyloxazole
PPO: 2,5-diphenyloxazole
POPOP: 1,4-bis-2-(4-methyl-5-phenyloxazolyl)-benzene
PB: 1,1',4,4'-tetra-phenyl butadiene
TPB: tri-*n*-butyl phosphate

Figure 1 (*17*) depicts the apparatus used by the Health and Safety Laboratory group in New York. The sample itself is in contact with the phosphor and covered with 0.005-inch Mylar® secured with Nylon® rings. The stability and the efficiency of counting are illustrated in table 2 (*17*) for calcium 45 ($0.25\beta_m$), tungsten 185 ($0.43\beta_m$), thallium 204 ($0.765\beta_m$), and yttrium 91 ($1.55\beta_m$).

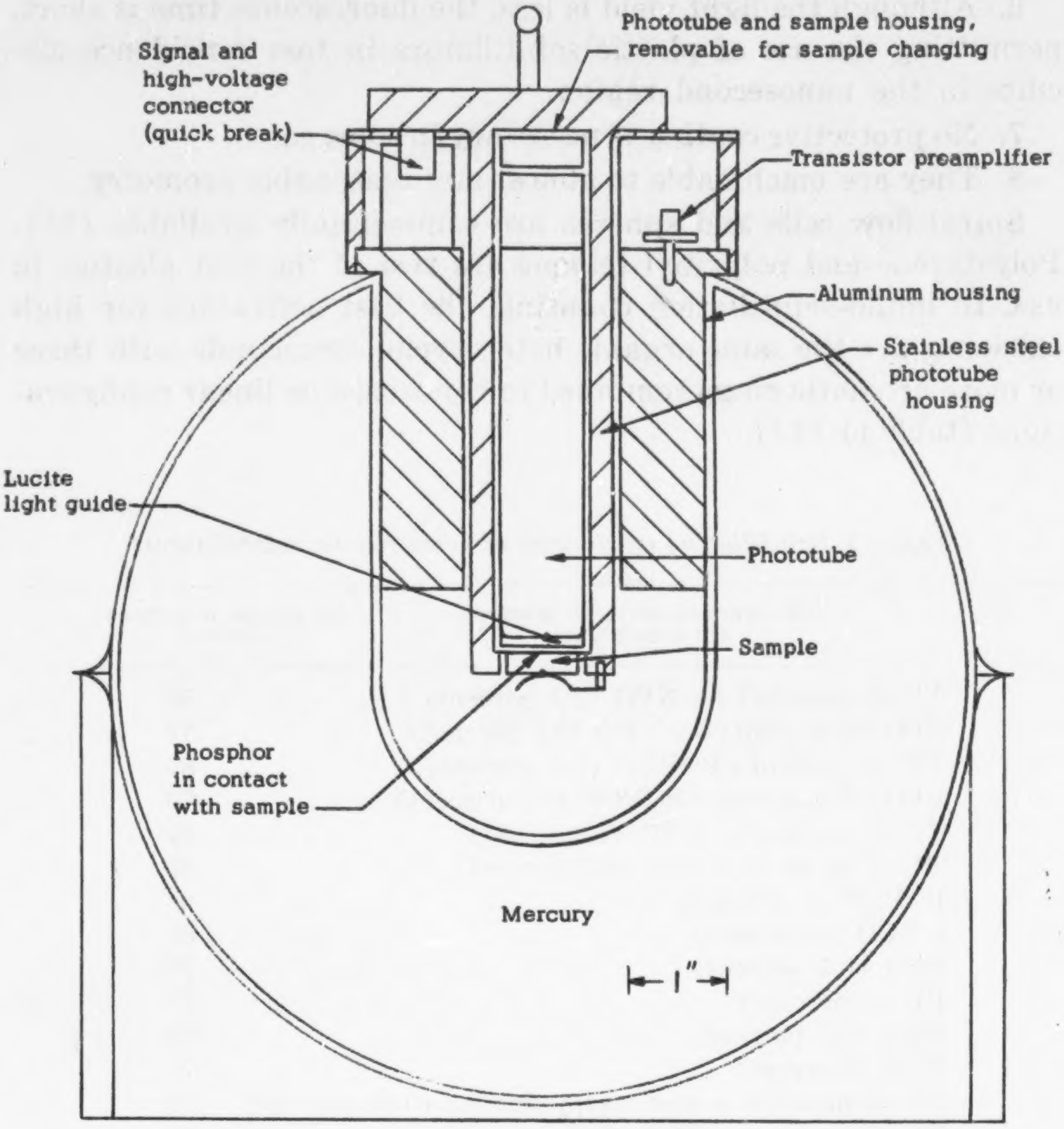

Phototube and sample housing, removable for sample changing
Signal and high-voltage connector (quick break)
Transistor preamplifier
Aluminum housing
Stainless steel phototube housing
Lucite light guide
Phototube
Sample
Phosphor in contact with sample
Mercury
1"

Figure 1. Beta-scintillation counter (*17*).

Figure 2 demonstrates that above 10 mils thickness there is no increase in efficiency. An anticoincidence umbrella can be used (*18*) to improve the figure of merit (fig. 3).

The plastic scintillator can be made very thin to reduce gamma-ray detection and background. Masking with a thin layer of aluminum foil filters out alpha particles if they are present in the sample, and furthermore the thin surface can exceed the maximum dimension of the opening of end-window counters.

If a thickness of about 1 centimeter is used, all the energy from the electron is absorbed and the amplitude of the pulse is approxi-

TABLE 2. *Efficiency runs for radionuclides* [a]

Day	Efficiency for calcium 45 (percent)	Efficiency for tungsten 185 (percent)	Efficiency for thallium 204 (percent)	Efficiency for yttrium 91 (percent)
1	21.8	23.8		49.1
2	22.0	23.6		48.7
3	21.9	24.3	37.3	48.9
4	21.4	23.8	36.9	47.2
5	21.7	24.0	37.3	47.3
6	21.6	24.0	35.1	47.1
7	21.7	24.3	36.8	47.4
8	21.8	24.2	37.2	48.0
9	21.9	24.4	36.8	48.7
Average efficiency	21.8	24.0	36.8	48.0
Measured standard deviation (Gaussian)	0.9	1.2	2.2	1.7
Expected standard deviation (Poisson)	0.5	0.7	0.4	0.5

[a] Reference (*17*).

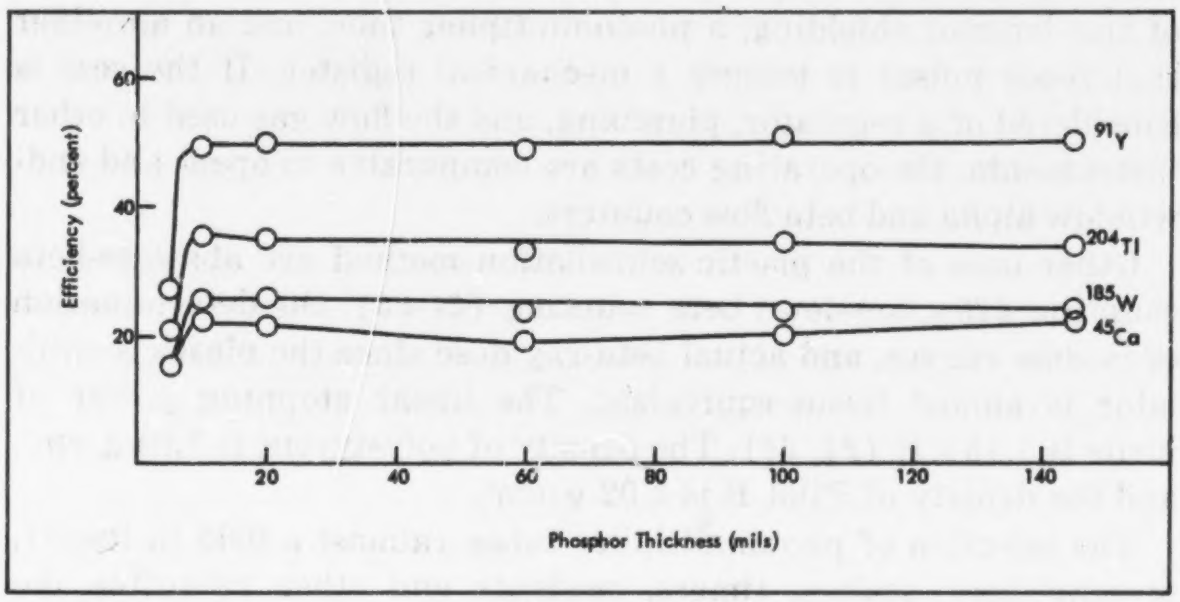


Figure 2. Plots of efficiencies against phosphor thicknesses.

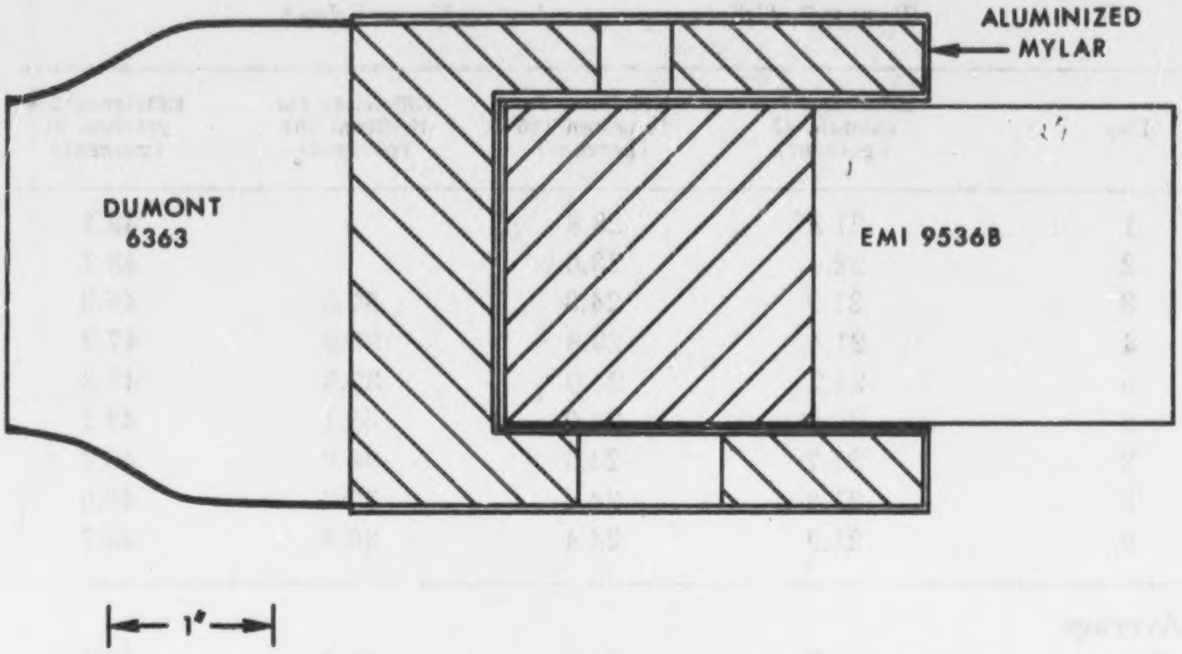


Figure 3. Schematic diagram of the β-ray spectrometer.

mately proportional to the energy of the beta particle. It is possible to discriminate against low energy and noise pulses or to determine the spectrum by feeding the pulses into a multichannel analyzer (*18, 19*). If one uses beta spectroscopy, radiochemical methods can be shortened or eliminated where two or more beta emitters are present.

A commercial version of the Health and Safety Laboratory instrument is now available. Specifications state background to be approximately 1 count per minute, which is adequate for low-level environmental radioassay. It is a simple, portable instrument that uses the disposal plastic disc scintillators (fig. 4). It consists of the detector shielding, a photomultiplier tube, and an amplifier that feeds pulses to trigger a mechanical register. If the cost is considered of a regulator, plumbing, and the flow gas used in other instruments, the operating costs are comparable to open- and end-window alpha and beta flow counters.

Other uses of the plastic-scintillation method are absolute-beta counting (*20*), low-level beta counting (*21-23*), the determination of isodose curves, and actual beta-ray dose since the plastic scintillator is almost tissue-equivalent. The linear stopping power of lucite is $1.15 \times H$ (*24, 25*). The density of polystyrene is 1.05 g/cm^3, and the density of Pilot B is 1.02 g/cm^3.

The selection of photomultiplier tubes (almost a field in itself), preamplifiers, scalers, timers, readouts and other recording devices, voltage supplies, and analysers is often peculiar to the re-

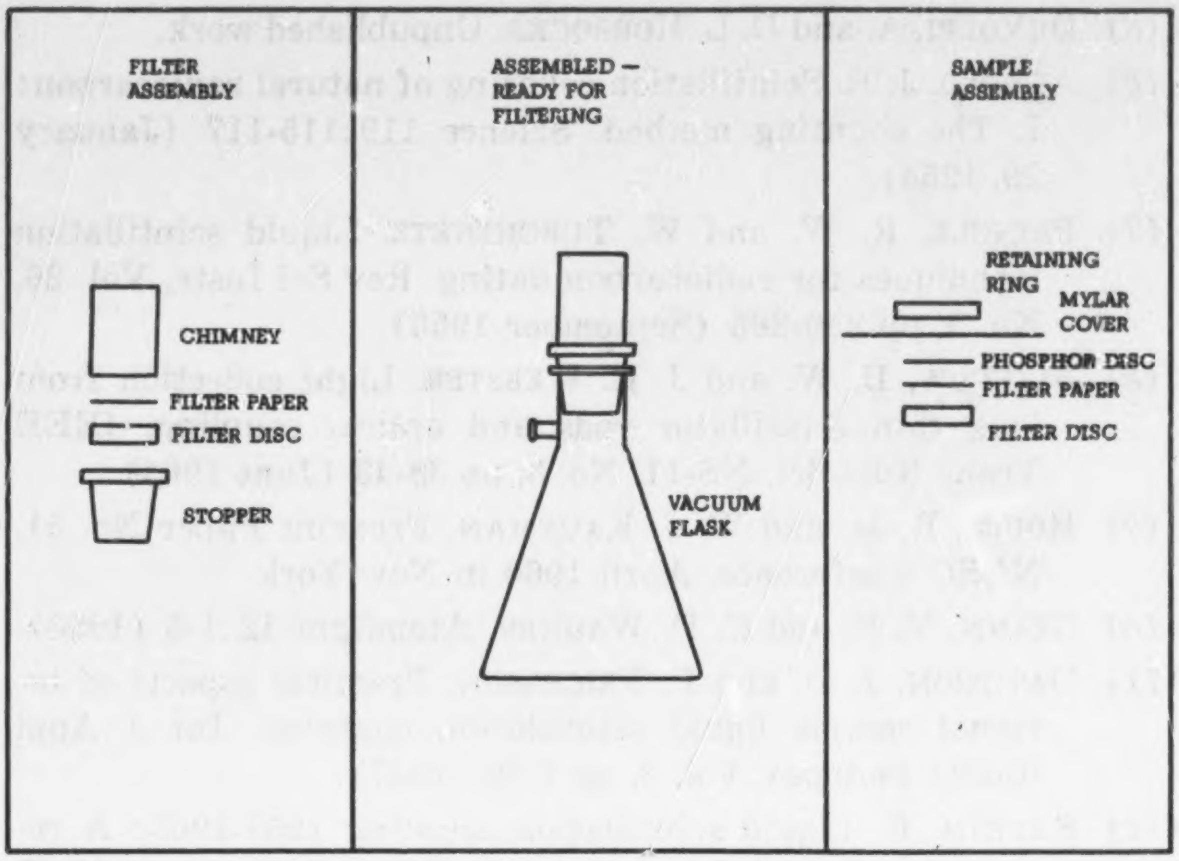

FILTER ASSEMBLY
ASSEMBLED – READY FOR FILTERING
SAMPLE ASSEMBLY
CHIMNEY
FILTER PAPER
FILTER DISC
STOPPER
VACUUM FLASK
RETAINING RING
MYLAR COVER
PHOSPHOR DISC
FILTER PAPER
FILTER DISC

Figure 4. Sample preparation.

sults desired, and perhaps the characteristics of the type of research or surveillance under consideration. Instruments and detectors should be housed in temperature- and humidity-controlled rooms. The voltage should be regulated and the room should be free of contamination and "electronic noise."

REFERENCES

(*1*) HORROCKS, D. L. Liquid Scintillation Counting of Inorganic Radioactive Nuclides. Packard Technical Bulletin 2:1-18 (Revised 1962).

(*2*) RONZIO, A. R. Int J Appl Radiat Isotopes 4:196 (1959).

(*3*) HELF, S., C. G. WHITE, and R. N. SHELLEY. Radioassay of finely divided solids by suspension in a gel scintillator. Anal Chem, Vol. 32, No. 2, pp 238-241 (February 1960).

(*4*) ERDTMANN, VON G. and G. HERRMAN. Z Elektrochem 64:1092 (1960).

(*5*) **DeVolpi**, A. and D. L. **Horrocks**. Unpublished work.

(*6*) **Arnold**, J. R. Scintillation counting of natural radiocarbon: I. The counting method. Science 119:115-117 (January 29, 1954).

(7) **Pringle**, R. W. and W. **Turchinetz**. Liquid scintillation techniques for radiocarbon dating. Rev Sci Instr, Vol. 26, No. 9, pp 859-865 (September 1955).

(*8*) **Blincow**, D. W. and J. R. **Webster**. Light collection from long thin scintillator rods and optical coupling. IEEE Trans Nucl Sci, NS-11, No. 3, pp 38-43 (June 1964).

(*9*) **Hours**, R. M. and W. J. **Kaufman**. Preprint Paper No. 51, NESC Conference, April 1960 in New York.

(*10*) **Guinn**, V. P. and C. D. **Wagner**. Atomlight 12:1-5 (1960).

(*11*) **Davidson**, J. D. and P. **Feigelson**. Practical aspects of internal sample liquid scintillation counting. Int J Appl Radiat Isotopes, Vol. 2, pp 1-18 (1957).

(*12*) **Rapkin**, E. Liquid scintillation counting 1957-1963: A review. Int J Appl Radiat Isotopes, Vol. 15, No. 2, pp 69-87 (February 1964).

(*13*) **Shorr**, M. and F. **Torney**. Physiol Rev 80:474 (1950).

(*14*) U.S. **Atomic Energy Commission**. Scintillation method in radiometry, AEC-Tr-5259. U.S. Atomic Energy Commission, Washington, D.C. (1961).

(*15*) **Raleigh**, H. D. and R. L. **Scott**. Nuclear Instrumentation, TID-3550, p. 99. U.S. Atomic Energy Commission, Washington, D.C. (1960).

(*16*) **Funt**, B. L. and A. **Hetherington**. Spiral capillary plastic scintillation flow counter for beta assay. Science, Vol. 129, No. 3360, pp. 1429-1430 (May 22, 1959).

(*17*) **Harley**, H., N. A. **Hallden**, and I. M. **Fiseene**. Beta scintillation counting with thin plastic phosphors. Nucleonics, Vol. 20, No. 1, pp. 59-61 (January 1962).

(*18*) **Kennett**, T. J. and G. L. **Keech**. An anti-coincidence β-ray scintillation spectrometer. Nucl Instr Methods, Vol. 24, No. 2, pp 142-148 (August 1963).

(*19*) **Shaskin**, V. L. Methods for analysis of natural radioelements. AEC-Tr-5458, State Publishing House, Moscow, Russia (1961).

(*20*) **Radaszewski**, T. and N. G. **Trott**. Phys Med Biol 7:3, p 319 (1962).

(*21*) Cross, W. G. AECL-802, Paper 211. U.S. Atomic Energy Commission, Washington, D.C. (1960).

(*22*) Becker, D. A. NBS Technical Note 284:75-77 (July 1963 to June 1964).

(*23*) Snyder, R. E. and G. B. Beard. A 4-pi beta spectrometer. Nucl Instr Methods 26:31-33 (February 1964).

(*24*) Goodwin, P. N. Scintillation counter for beta-ray dosimetry. Nucleonics, Vol. 14, No. 9, pp. 120-123 (September 1956).

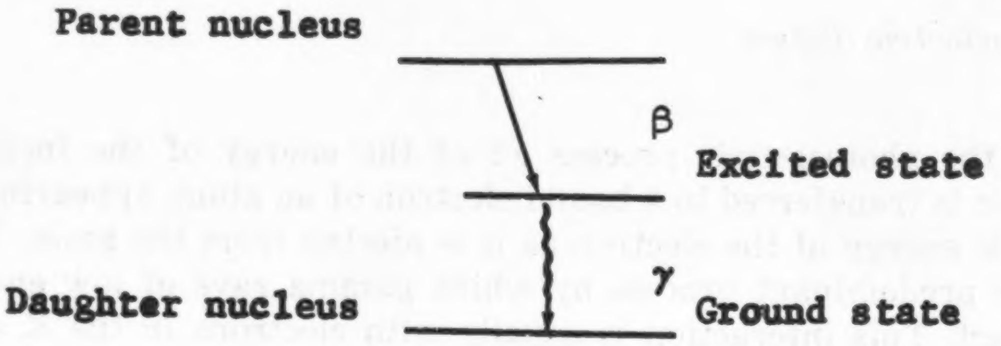
(*25*) Chhabra, A. S. Sr^{90}-Y^{90} beta-ray (and Bremsstrahlung) depth-dose measurements in lucite. Radiology, Vol. 79, No. 6, pp 1001-1007 (December 1962).

Chapter III. Pulse-Height Analyzers and Gamma-ray Spectrum Analysis [1]

Allan E. Smith [2] and *Robert W. Coulter* [3]

Gamma rays are electromagnetic radiation resulting from the transition of a nucleus between one discrete energy level and another. Because these transitions occur between discrete levels, the emitted gamma rays (photons) from a given radionuclide have discrete characteristic energies. (Although these gamma rays are characteristic of a given radionuclide, they are not necessarily unique. Several radionuclides may emit photons with the same energy.)

The emission of a gamma ray usually follows decay by alpha- or beta-particle emission, and this leaves the nucleus in an excited state:



It is not uncommon for this decay pattern to result in the emission of many (occasionally 10 or more) gamma photons for a given parent radionuclide depending on the energy of the alpha or beta emission. Other types of decay may produce direct gamma emission without the prior emission of alpha or beta particles (for example, electron capture as in the case of strontium 85 and isomeric transition as in the case of barium 137m).

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[1] Much of this descriptive material has been condensed from the manual for the course "Radionuclide Analysis by Gamma Spectroscopy," which is presented several times each year by the Training and Manpower Development Program, National Center for Radiological Health, Rockville, Md. 20852.

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PROCESS OF DETECTION

Interaction of Gamma Rays with Matter

The detection of gamma rays depend on their interaction with the detecting medium (for the purposes of this section—NaI) in some manner to produce ultimately an electrical pulse at the output of the electronic multiplier. The three main processes by which photons transfer all or part of their energy to the detecting medium are (1) photoelectric effect, (2) Compton scattering, and (3) positron-electron pair production. The probability of interaction by each of these processes will vary with the energy of the incident photon and the atomic number of the absorbing medium.

Photoelectric Effect

In the photoelectric process all of the energy of the incident photon is transferred to a bound electron of an atom, appearing as kinetic energy of the electron as it is ejected from the atom. This is the predominant process by which gamma rays of low energy interact. This interaction is usually with electrons in the K shell of the atom as long as the incident photon energy exceeds the binding energy for the K shell. As a consequence of the ejection of an electron, the atom is left with a vacancy in the orbital shell. As some other electron falls into this vacancy, x rays or Auger electrons are emitted. Since this process occurs in a short period of time relative to the other processes involved in scintillation, the secondary x rays may also interact with the crystal by the photoelectric process, resulting in complete absorption of all of the energy of the incident photon with the scintillator. Assuming an idealized spectrometer system, the net result of this process alone would be the production of a line spectrum where all incident photons that interact would be represented by a single line at the appropriate energy. The fact that this does not occur is the sum result of the effects of the nonproportional response of the crystal, the statistical variation in the scintillation process and the electron multiplication by the phototube, and the small angle backscatter from the crystal container. When the incident photon has an energy much greater than that required to eject an electron, the photoelectric process becomes less probable.

Compton Scattering

In the process of Compton scattering, incident photons interact with electrons that are essentially free and undergo a partial energy loss. The energy of the incident photon is shared between the electron and scattered photon. The energies of the scattered photon and electron are given by:

$$E'_\gamma = \frac{E_\gamma}{1+a(1\text{-}\mathrm{Cos}\theta)} \qquad \left[a=\frac{E_\gamma}{mc^2}\right]$$

$$\text{and } E_e = E_\gamma - \frac{E_\gamma}{1+a(1\text{-}\mathrm{Cos}\theta)}$$

where E_γ is the incident gamma ray energy,
E'_γ is the energy of the scattered gamma ray,
E_e is the scattered electron energy,
m is the rest mass of the electron,
c is the speed of light,
mc^2 is equal to 0.511 MeV, and
θ is the angle between the directions of the incident and scattered photons.

The resulting Compton electron energy spectrum will extend from zero energy ($\theta=0°$) to the maximum value that is slightly less than the energy of the incident photon ($\theta=180°$). The maximum energy of the Compton electron (E_C) is given by:

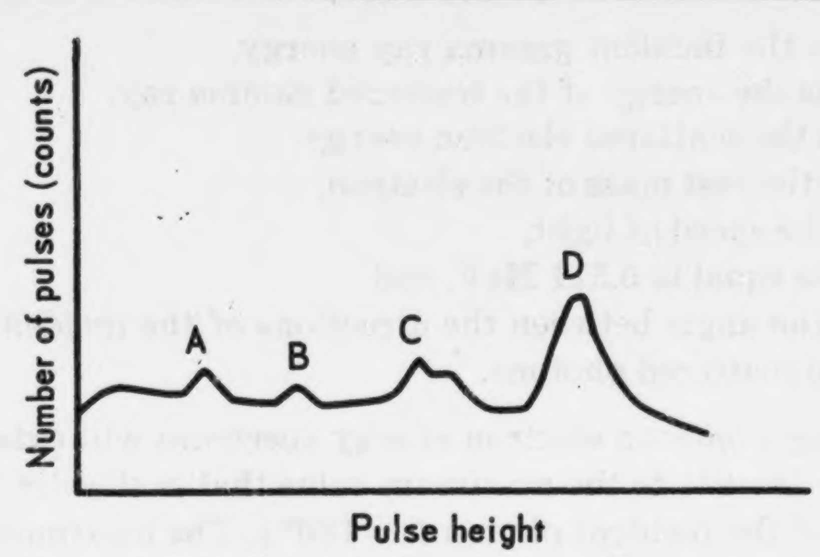
$$E_C = E_\gamma - \frac{E_\gamma}{1+2a}.$$

This energy represents a sharp cutoff of the Compton continuum and is referred to as the Compton edge; thus it can be seen that the contribution from Compton scattering can affect the entire spectrum below a given photopeak. Following this process, the Compton electron will lose energy through secondary ionization, and the scattered photon may undergo further Compton interactions, may lose all of its energy through a photoelectric interaction, or may escape the detecting medium.

Pair Production

For incident gamma rays having an energy in excess of 1.02 MeV, interaction resulting in the production of a positron-electron pair is possible. (For practical purposes, pair production becomes significant for incident photons in the neighborhood of 2 MeV or greater.) In the process of pair production, the incident photon disappears and the total energy of the particles will be equal to the energy of the incident photon, with their kinetic energy equal to the total energy minus their rest energy (2 mc^2 or 1.02 MeV).

Since the positron is unstable in the presence of an electron, annihilation occurs with the emission of two photons of 0.511 MeV (energy equivalent of the rest mass of the particles). There will be a net energy loss in the detector equal to the incident photon energy minus 1.02 MeV. It is possible for either or both of the 0.511 MeV photons to interact with the detector by either the photoelectric or Compton process (fig. 1).



A. 0.511 MeV (annihilation energy - detection of photons from pair production external to the detector).
B. Full energy - 1.02 MeV (both photons escape).
C. Full energy - 0.511 MeV (one photon escapes).
D. Full energy photopeak.

Figure 1. Illustration of full-energy escape and annihilation peaks.

This series of events can result in energy loss to the detector of energies ranging from that of the incident photon down to the energy of the incident photon minus 1.02 MeV; however, the probability of interactions at certain energies is sufficiently high to produce a series of secondary photopeaks characteristic of the pair production process.

Related Spectrum Components

In addition to the processes previously described by which gamma rays interact with the detector and lose all or part of their energy, certain related interactions may contribute to the overall shape and characteristics of the gamma spectrum.

Backscatter Peak

The backscatter peak arises from the Compton scattering process and is superimposed on the Compton continuum. It results from the scattering, through approximately 180°, of photons that have interacted with materials surrounding the detector, such as the "shield," and the detection of these degraded photons by the detecting medium. The energy of this backscatter peak may be approximated from the general Compton energy equation assuming $\theta = 180°$. This reduced equation becomes:

$$\text{Energy of backscatter peak} = \frac{E_\gamma}{1 + 3.92\, E_\gamma}$$

As the energy of the incident photon increases, the energy of the backscatter peak is limited to a maximum of 0.255 MeV.

Annihilation Peaks

When the pair production process occurs external to the detector, there is a possibility that one of the 0.511 MeV photons that results will enter the detector and lose all of its energy. When this occurs a photopeak may occur at a pulse height corresponding to 0.511 MeV. This is represented by the annihilation peak in figure 1.

Sum Peaks

Sum peaks result from the interaction of more than one photon with the detector in a short period of time. These multiple events are seen as a single interaction by the detector, and a photopeak equal to the sum of the energies of the individual photons will result. This is more likely to occur in samples of high activity.

Common sources of sum peaks are those isotopes that emit gammas in cascade, such as cobalt 60, where the 1.17- and 1.33-MeV photons can "sum" to produce a photopeak at 2.50 MeV.

Components of the Gamma Spectrometer

A gamma-ray spectrometer is a combination of a scintillation detector, which will produce electrical pulses representative of the energy of gamma photons incident on the detector, and a pulse-height analyzer capable of distinguishing incident photon energies through analysis of electrical pulses of different magnitude and determining the number of pulses occurring at each magnitude. The data obtained from the spectrometer are in two-dimensional form, usually expressed as count rate versus energy.

In turn these spectral data can be interpreted or analyzed to determine which radionuclides contribute to the spectrum (qualitative analysis) and how much activity of each component radionuclide there was (quantitative analysis).

The components of a gamma-spectrometer system can be grouped into three basic functional systems as indicated in figure 2:

1. The detector system composed of a scintillation detector assembly, preamplifier, detector high-voltage power supply, and shield.
2. The pulse-analyzer system composed of the linear amplifier, pulse-height analyzer, memory storage, and control logic.
3. The readout system containing one or more readout devices includes oscilloscope, readout indicator, electric typewriter, digital printer, paper tape perforator, magnetic tape recorder, strip chart recorder, and X-Y recorder.

These components will be presented only in relation to their function.

DETECTOR SYSTEM	PULSE-ANALYZER SYSTEM	READOUT SYSTEM
• SCINTILLATION DETECTOR ASSEMBLY • PREAMPLIFIER • DETECTOR HIGH-VOLTAGE POWER SUPPLY • SHIELD	• LINEAR AMPLIFIER • PULSE-HEIGHT ANALYZER • MEMORY STORAGE • CONTROL LOGIC	• OSCILLOSCOPE • READOUT INDICATOR • ELECTRIC TYPEWRITER • DIGITIAL PRINTER • PAPER TAPE PERFORATOR • MAGNETIC TAPE RECORDER • STRIP CHART RECORDER • X-Y RECORDER

Figure 2. Gamma-spectrometer components.

The Detector System

Scintillation Detector Assembly

The scintillation detector assembly consists of three basic components, including scintillation crystal, photomultiplier tube, and mu-metal shield, all incorporated within a light-tight assembly. In some systems, the preamplifier also may be incorporated within the assembly.

The function of the scintillation crystal is to convert the energy of the incident gamma photons to light pulses. The amount (or intensity) of light produced is directly proportional to the energy transferred to the crystal when a gamma photon interacts with it. For each gamma interaction there is produced one flash of light that is representative of that interaction. The crystal is usually "canned" in an aluminum, stainless steel, or copper container. An optical coupling compound is used between the crystal and photomultiplier tube to improve light transmission.

The light energy produced in the crystal is converted into electrical energy by means of the photomultiplier tube, which is composed of the photocathode and the electron multiplier. Light striking the photocathode releases electrons, through the photoelectric effect, in proportion to the amount of light incident on the photocathode. The small number of electrons produced are multiplied by accelerating them across a series of voltage potentials striking successive targets (dynodes), each releasing more than 1 electron for each incident electron. The group of electrons produced from one gamma interaction forms a pulse whose height is proportional to the energy transferred in the original interaction.

A mu-metal shield is placed around the photomultiplier tube to reduce magnetic effects from the earth and the steel shield. The entire crystal detector assembly must be light-tight.

Preamplifier

A preamplifier is used to condition the low-level pulse emitted from the photomultiplier tube for delivery to the linear amplifier. Without conditioning, the pulse would be severely attenuated by long cable runs between the detector system and the pulse-analyzer system. A preamplifier is not always necessary. It may be incorporated within the pulse-analyzer system; however, some form of signal conditioning is necessary at the photomultiplier tube to produce a signal acceptable to the pulse-analyzer system.

Detector High-Voltage Power Supply

A detector high-voltage power system is necessary to operate the photomultiplier tube. The supply should be capable of delivering a stable, noise-free potential of approximately 1,000 volts. A positive polarity is preferred by most photomultiplier manufacturers. The supply may be a separate part or a component part of the pulse-analyzer system.

Shield

The shield reduces the naturally occurring radioactive background so that the effect of statistical variations in the net count rate will be as small as possible. The shield will be discussed in greater detail later in this report.

The Pulse-Analyzer System

In Geiger-Mueller or proportional counting systems, the individual pulses from the detector are recorded on a scaler to indicate the total number of pulses, or counts, that have occurred in a given time period. The gamma spectrometer not only determines the number of pulses that have occurred, but also classifies them into given energy ranges. This classification is done by the pulse-analyzer system, which contains a preamplifier, linear amplifier, pulse-height analyzer, and storage unit.

Preamplifier and Linear Amplifier

These two units convert the weak, low-amplitude pulse emitted from the photomultiplier tube to a stronger, higher amplitude pulse that can be readily handled by the succeeding pulse-height analyzer. The two primary characteristics of these units are that they produce pulses of uniform shape and that the size of the pulse is in direct linear proportion to the pulse from the photomultiplier tube.

Pulse-Height Analyzer

The pulse from the linear amplifier is then fed into a pulse-height analyzer capable of distinguishing pulses of different predetermined magnitudes. The number of pulses of each magnitude is then recorded or stored.

Memory Storage

The storage unit is the filing system of the spectrometer. The information that is continuously generated in the pulse-height analyzer is added together and compiled as a running total of the number of pulses in each pulse-height range. At the end of the sample counting period, the information in the storage unit is a compilation of all the different pulses originating from the detector system during the counting period.

Control Logic

Control logic is necessary to handle the various data manipulations in an analyzer system. Control logic allows the operator to select various modes of operation at the input and output of the analyzer system.

Readout System

The pulse-analyzer system can present the stored data in various forms. In single-channel and earlier multichannel (voltage-discriminator type) analyzers, the storage and readout units were one and the same; namely, a scaler or a set of scalers. In present computer-type multichannel analyzers, the information in the storage unit is in a form that cannot be visually interpreted without the aid of a readout device. The data must be converted from one form to another to operate the various readouts. Some analyzers incorporate the converters within the basic analyzer system while others use separate units. The readout device must be compatible with the analyzer system.

Readout Devices

The various readout capabilities are listed below. Not all readouts are necessary or desirable for an analyzer system. Reference (*3*) provides a complete description of readouts.

OSCILLOSCOPE

A cathode-ray oscilloscope is usually a part of the analyzer system. The oscilloscope provides a visual graphic display of the data in the memory storage unit.

READOUT INDICATOR

A readout indicator provides a digital display of data stored in any single channel. It may be an accessory item and is not always needed.

ELECTRIC TYPEWRITER

An electric typewriter provides, in printed format, the digital data from the memory storage unit.

DIGITAL PRINTER

A digital printer also provides a printed format of the digital data and is generally faster than a typewriter.

PAPER TAPE PERFORATOR

Commonly called a "tape punch," the perforator provides a data format compatible for computer analysis. A tape perforator is not necessary unless automatic data anlaysis methods are used.

MAGNETIC TAPE RECORDER

A magnetic tape recorder is similar in function to paper tape and provides faster readout. Magnetic tape recorders are not necessary unless automatic data analysis methods are used.

STRIP CHART RECORDER

A strip chart recorder can be used to produce a permanent readout similar to the oscilloscope display. Roll chart paper is used.

X-Y RECORDER

An X-Y recorder can also produce a permanent readout similar to the oscilloscope display. Standard graph paper is used.

SYSTEM PERFORMANCE

The performance of an analyzer system from an operational point of view is described below and can be expressed in terms of two concepts: (1) resolution—the ability of a system to differentiate between gamma photons of different energies—and (2) lower limit of detection—the ability to determine with a given degree of certainty the lowest possible concentration of gamma-emitting radionuclides.

Resolution

The characteristic photopeaks of a gamma spectrum are the principal means by which qualitative and quantitative analyses are performed. The resolution of a system is used to describe these photopeaks in terms of width versus height and to define the minimum energy difference that can be distinguished.

Percent resolution is defined as the ratio between the energy interval in MeV at one-half maximum peak height and the photopeak energy. By common acceptance, cesium 137 is the radionuclide of reference for comparative purposes.

$$\text{Percent resolution} = \frac{E_{1/2} \times 100}{E}$$

where E is the photopeak energy in MeV and
$E_{1/2}$ is the photopeak width in MeV at one-half maximum height.

The percent resolution of a system is an intrinsic function of the detector-photomultiplier combination. Commercially available detector systems can be obtained with a guarantee of approximately 8 percent (for cesium 137) resolution for 4- by 4-inch NaI crystal assemblies.

Lower Limit of Detection

The ability of a system to define the lowest possible concentration of radioactivity in a sample is primarily a function of counting geometry (which affects efficiency) and the system background. Since counting geometry will be determined by the type and size of the samples to be counted, this will be covered together with calibration procedures in a later section.

The system background is the count rate when a sample containing no radioactivity is placed on the detector and includes contributions from the following sources:

1. Internal radioactivity—natural or manmade contamination of the detector system, such as the presence of potassium 40 in the crystal or photomultiplier tube.

2. External radioactivity—natural or manmade contamination, such as radium in lead shields, or in concrete walls of the counting room, and from cosmic radiation that penetrates the shielding around the detector.

3. Electronic noise—pulses originating in the electronic circuitry of the system.

To reduce background, contributions from each of these sources must be minimized.

INTERNAL RADIATION

Although it may not be possible to eliminate internal radioactivity completely, it can be greatly reduced. Specifications that call for low potassium (2 ppm) crystals and photomultiplier tubes can be met easily and will serve to reduce the potassium 40 contribution at very little increase in price. Similarly, casings for the crystal should be specified as being free of manmade contamination. Aluminum casings are more apt to have slight amounts of natural radioactivity than stainless steel or copper.

EXTERNAL RADIATION

The largest contributors to background levels are the external radiation because of cosmic rays, radon daughter products in the air, radionuclides emitting gamma rays in the vicinity of the detector, and the contamination of building materials by natural radioactivity.

1. Cosmic radiation, composed mostly of high-energy mesons and nuclei, cannot be shielded against effectively. Primary interference will occur well above the energy range of interest for analysis of environmental samples, and the increase in background will not be great. Lower energy cosmic secondaries can be shielded against with some success by massive shielding (4 to 6 inches of lead or steel). "Hard" cosmic rays will interact with the shielding to produce secondaries within the shield. The net result is very little reduction in cosmic rays.

2. Radon gas and radon daughter products are natural contaminants of the atmosphere. Their concentration in ambient air will

vary according to temperature, humidity, and other weather conditions, and may reach levels sufficient to cause major changes in background levels. Radon daughter products can be filtered from the air to prevent their buildup; radon itself emits no gamma radiation. While a closed system of aged, filtered, and recirculated air is most effective, it may involve the installation of an extensive circulating and filtering system not readily adaptable to the physical facility. A satisfactory method, which is much simpler and less expensive, involves filtering outside air at a high rate of turnover.

3. Building materials often contain natural gamma-emitting radionuclides such as radium, thorium, and uranium. Massive shielding can eliminate the contribution of these gamma emitters to the background.

MASSIVE SHIELDING

The use of massive lead or steel shielding (4 to 6 inches thick) is the most effective means of reducing background radiation. The design and construction of such a shield require consideration of several factors.[4] Limitations, because of size and weight capacity of the proposed counting room, may dictate certain requirements.

1. Thickness of shield—gamma-ray transmission will vary approximately as e^{-x} where x is the shield thickness. Increased shielding does not indefinitely reduce gamma transmission to zero; however, a maximum effect is found with 6 to 7 inches of steel shielding. Thickness in excess of this does not significantly reduce the background further. Denser materials may be substituted to reduce wall thickness.

2. Shielding material—table 1 provides comparisons between various shielding materials. The half-value thickness (HVL) is that thickness required to reduce gamma intensity to one-half of its incident value.

The material used in construction must be free of radioactive contamination. This can be written into specifications, or the material may be checked by using an existing system.

3. Size (internal) will determine to some extent the backscatter characteristics of the shield. Backscatter will be more severe the closer the interior walls are to the detector. The internal dimen-

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[4] Sample specifications are included in appendices.

sions should be as large as cost, weight, and space will permit. A practical compromise is represented by inner side dimensions, on the order of 24 to 36 inches.

4. Internal liners can also be used to reduce the backscatter because of Compton interactions. A lead liner of one-eighth inch in a steel shield will serve to significantly reduce backscatter through absorption of the photons scattered by the walls of the shield. It will in turn give rise to lead x rays, which can be reduced by the use of a second liner of one-sixteenth inch of cadmium.

In lead shields the x-ray problem predominates and liners are used that have high cross-section for the absorption of fluorescent radiation. A series consisting of one-sixteenth inch of cadmium followed by one-sixteenth inch of copper is commonly used. Many other materials have been used successfully, including tin and lucite.

TABLE 1. *Shielding properties of common materials for 2.0-MeV gamma rays*

	Density (g/cm^3)	(HVL) (inches)	Equivalent thickness (inches Fe)	μ (cm^{-1})	μ/density (cm^2/g)
Water	1.00	5.45	6.50	0.050	0.050
Concrete	2.35	2.68	3.00	.106	.045
Steel	7.80	0.82	1.00	.335	.043
Lead	11.34	.53	0.57	.510	.045
Mercury	13.54	.44	.48	.624	.046

CALIBRATION PROCEDURES

Energy Calibration

The analog display and digital printout of the data accumulated by a pulse-height analyzer represent the total number of counts collected in a given time period by channel number. Since analysis of all data depend on the identification of the various radionuclides by their full-energy photopeaks, the channel number is meaningless until an energy-channel-number relationship has been defined.

The desired relationship is a proportional one where the energy can be defined from $y = mx$: energy = channel coefficient X channel number. The channel coefficient is equal to incident gamma energy divided by the channei number and may be adjusted by varying the analyzer gain.

A second type of relationship that may exist between channel number and incident photon energy is the linear relationship expressed by $y = mx + b$ (energy = channel coefficient X channel number + constant).

These two types of response are illustrated by figure 3.

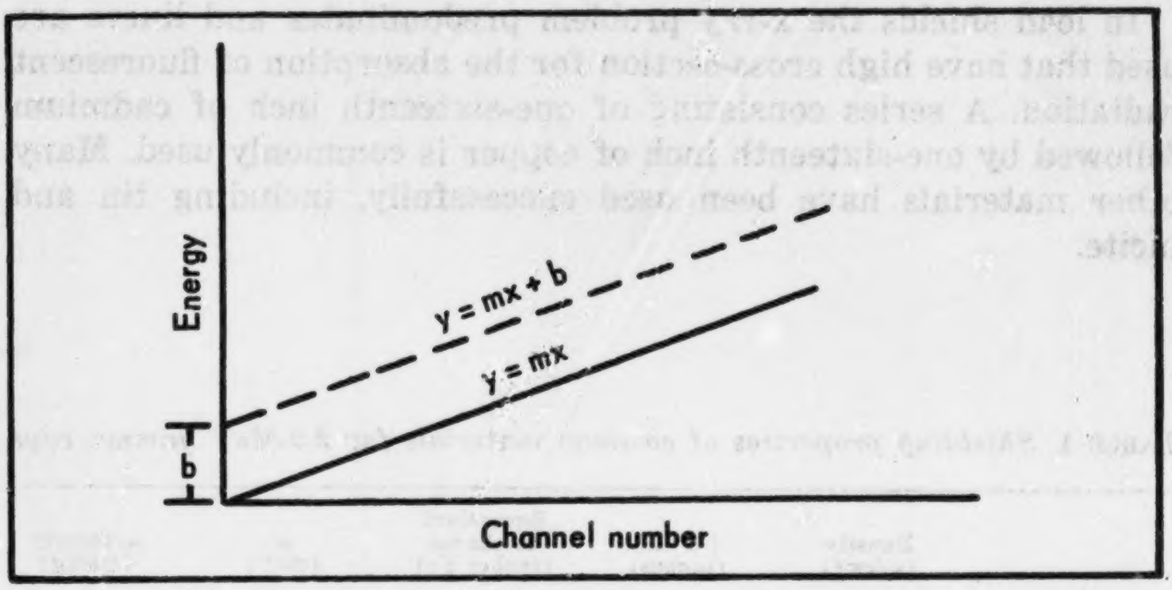

Figure 3. Two idealized types of linear response of pulse-height analyzers.

Although the proportional response is desired, it is not always easily obtained. In practice, much of the curve will be almost linear (but not necessarily all of it), and it may not intercept at a gamma energy of zero. The proportionality (or linearity) of an analyzer system is dependent on the detector and the electronic components of the analyzer (particularly the amplifier). Exact linearity cannot be obtained since the response of sodium iodide crystals is not exactly linear.

Selection of Channel Coefficient

Since the purpose of obtaining proportional response curves is to provide a constant channel coefficient for given analyzer settings (gain, high voltage, zero level, and so forth), considerable thought should be given to its selection. The minimum value will be determined by the energy range to be examined. For environmental samples, a maximum energy between 2 MeV and 4 MeV is acceptable. A coefficient, which is easily used (for example, 0.005, 0.01, 0.02 MeV per channel), is desirable for routine operations. Excessively high coefficients make it difficult to differentiate between photopeaks of similar energies. A value selected can be achieved by varying the gain control to locate known peaks in given channels (for example, cesium 137, 0.662 MeV in channel 66).

Measurement of System Linearity

In describing the linearity of a system, reference is usually made to three types of linearity. These are usually quoted in manufacturers' specifications and can serve as a guide to the selection of an analyzer system.

1. Absolute linearity is the ratio of the error in the apparent energy to the value of the true energy. Since distortions may occur in different portions of the linearity curve, this must be expressed relative to the true energy.
2. Integral linearity is the ratio of the error in apparent energy to the full-energy scales. This factor will also depend on the energy at which the error is measured (as in 1 above). The value for the full energy scale will be the product of the channel coefficient times the number of channels available (the theoretical energy value of the last channel).
3. Differential linearity is a measure of the uniformity of the channel widths and is expressed as a percent maximum deviation from the average width.

The linearity of the system as a unit can be checked by measuring a series of radionuclides having gamma rays of known energies and plotting the energy of each photopeak by channel number. This will serve as a check on the total system including the detector.

A pulse generator may be used to check the linearity of the analyzer portion of the system by providing data for an energy versus channel number plot. (Many analyzers include built-in pulse generators.) By this method, the effect of the detector is eliminated since the pulse is routed directly into the analyzer, bypassing the detector.

The differential linearity of a system may be measured by employing a sweeping pulse generator that provides pulses of successively higher amplitudes at a constant frequency. As the amplitudes increase, the pulses will be assigned to successively higher channel numbers. The number of pulses accumulated in each channel will provide a measure of the channel width. A differential linearity of 1 to 2 percent is desirable.

Causes of Nonlinearity

The nonlinear conversion of gamma rays into light photons within the crystal will be reflected in the total system performance. The amount of light produced per unit energy of the incident photon is not a constant but will vary as a function of energy (particularly in the 0 to 2 MeV range). Above 0.2 MeV, this effect can be considered negligible.

The electronic circuitry of the analyzer itself may provide a second source of system nonlinearity. The amplifier, which should amplify pulses from the detector in a linear fashion, may not exhibit a constant gain over the entire energy spectrum. This distortion is most likely to occur at the higher energies.

Correction of Nonlinearity

If a linearity plot of the system reveals excessive deviations from the desired proportional response, some compensation may be made by shifting the zero channel of the analyzer so that it represents some channel other than zero. In general, this is not particularly desirable, and care should be taken to specify systems designed to yield linearity curves approaching a proportional response when coupled to the detector.

The analyzer system may be expected to "drift" (change energy calibration) slightly as a function of temperature and count rate with passing time. To help control or eliminate this drift, it is advisable to maintain temperature control within the counting facility. A temperature range of ±5° F. provides sufficient control and is usually not difficult to maintain. Daily checks on the value of the channel coefficient and linearity should be made by using a gamma source with two or more photopeaks. Cesium 137 and cobalt 60 together make a convenient check source having three photopeaks and a relatively long half-life. Adjustments in the fine gain control will normally return the analyzer to its previous calibration. If temperature fluctuations in a given day exceed those recommended above, more frequent checks are advised.

The temperature of the sample is a more important consideration in terms of possible damage to equipment than is temperature of the environment.

Since the sample will be in direct contact with the crystal detector, extremes in temperature of the sample that can cause cracking of the detector must be avoided. Damaged crystals show poor resolution and usually reduced efficiencies.

EFFICIENCY CALIBRATION

The determination of counting efficiencies for an analyzer system is the first step in the process of quantitative analysis. Since all subsequent analytical results will depend on this process, extreme care should be exercised in selecting and preparing the standards to be used. Several sources of supply for calibrated standards are available, and lists of radionuclides and services offered are provided. Among these are:

1. The Radiochemical Center, Amersham, Buckinghamshire, England.
2. Oak Ridge National Laboratory, Oak Ridge, Tenn.
3. National Bureau of Standards, Gaithersburg, Md.
4. Private firms specializing in nuclear instrumentation and services.
5. Analytical Quality Control Service, DRH.[5]

Standards supplied by these organizations will normally be shipped in flame-sealed glass ampoules. Certain radionuclides may tend to deposit on plastic containers; therefore the use of glass for dilution and storage is recommended. To avoid the possibility of loss because of plating of the radionuclides on glass containers, an isotopic carrier may be added to the solution.

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[5] The Division of Radiological Health was reorganized January 1, 1967. It is now the National Center for Radiological Health, with Mr. James G. Terrill, Jr., as Center Director. The Analytical Quality Control Service is located at the Northeastern Radiological Health Laboratory, Winchester, Mass. 01890.

INSTRUMENT CALIBRATION

Once the appropriate standards have been obtained, the calibration procedure for determining detection efficiencies is relatively straightforward. The chosen standards are diluted to the appropriate level, placed in the containers for the selected geometry, and a spectrum is taken. Several general considerations should be mentioned.

Selection of Configuration (Geometry)

The method of holding the sample (geometry employed) will be determined by the types of samples analyzed. For environmental samples three basic geometries probably will be most appropriate.

Marinelli Beaker

The Marinelli beaker (fig. 4) is most useful for liquid samples of fairly large volume. By varying the dimensions of the container, they can be designed to hold varying volumes depending on the desired size of the sample. These containers are useful for counting food samples that have been blended into a slurry (by the addition of distilled water if necessary).

Containers

Small plastic containers (such as those used to hold cottage cheese) are often useful for liquid or semisolid samples of small volume (approximately 400 ml). They can be utilized for solid materials such as mud or soil, but compensation for self-absorption effects may have to be made.

Extended Disc (Planchet)

Extended disc geometries of varying sizes may also be useful for counting samples that have been chemically separated, air

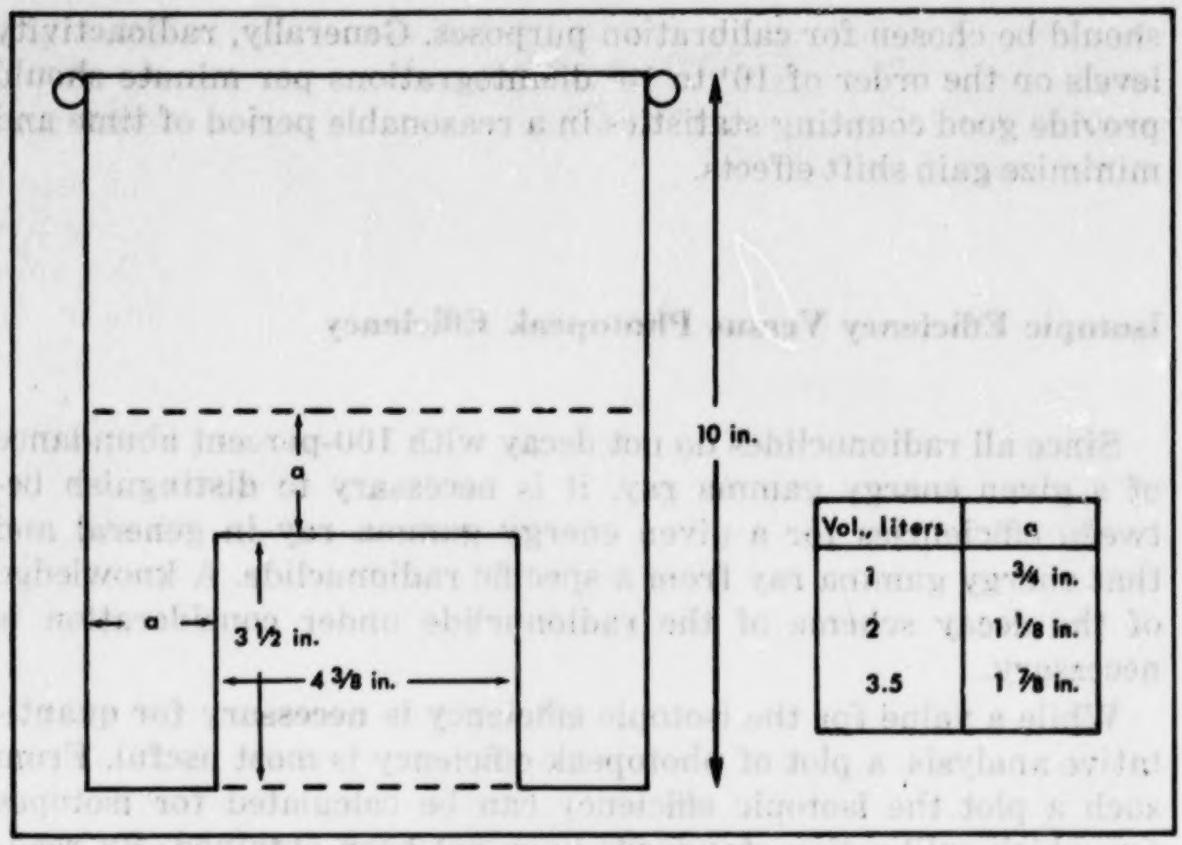


Figure 4. Cross section of special (Marinelli) aluminum beakers (three sizes).

filter samples, or small volume evaporated liquid samples. If a 4-inch-diameter crystal is employed, a 4- to 5-inch planchet could be used. This size conveniently holds 4-inch-diameter air filter samples.

Point Sources

A calibration for point sources (small diameter relative to crystal diameter) finds little application to environmental samples, but may be useful for secondary calibration of gamma-emitting radionuclides in solution.

Radioactivity Level

As indicated above, changes in radioactivity level may result in gain shifts in energy calibration of the analyzer. For this reason, standards that approximate the level of radioactivity expected should be chosen for calibration purposes. Generally, radioactivity levels on the order of 10^4 to 10^5 disintegrations per minute should provide good counting statistics in a reasonable period of time and minimize gain shift effects.

Isotopic Efficiency Versus Photopeak Efficiency

Since all radionuclides do not decay with 100-percent abundance of a given energy gamma ray, it is necessary to distinguish between efficiencies for a given energy gamma ray in general and that energy gamma ray from a specific radionuclide. A knowledge of the decay scheme of the radionuclide under consideration is necessary.

While a value for the isotopic efficiency is necessary for quantitative analysis, a plot of photopeak efficiency is most useful. From such a plot the isotopic efficiency can be calculated for isotopes for which calibration standards have not been obtained. By reading the photopeak efficiency of the gamma-ray energy involved and multiplying by the percent abundance from the decay scheme, a calculated value for the isotopic efficiency can be found; that is, Isotopic Efficiency = Photopeak Efficiency X Gamma Abundance.

Parent-Daughter Relationships

Several radionuclides commonly encountered in environmental samples (barium-lanthanum 140, zirconium-niobium 95) may exist in some degree of equilibrium with their daughter products. In these cases, radioactivity values can be calculated for one radionuclide or for both members of the pair.

QUALITATIVE ANALYSIS

The first step in the analysis of a gamma spectrum is an identification of the gamma emitters contributing to that spectrum. Of primary interest in this process will be the definition of the photopeak energies represented in the spectrum.

When applying the channel coefficient for the given system, an ordered list of photopeaks by energy can be made. It is wise to consider each photopeak in terms of an energy span plus-or-minus one channel on either side of the apparent peak to compensate for possible system drift. Reference can then be made to a table of radionuclides by gamma-ray energies to provide a list of possible radionuclides for each energy. Since certain regions of the spectrum (particularly the low energies) offer a great many possibilities, further refinement will be necessary to reduce the number of radionuclides under consideration. To allow for the possibility of overlapping photopeaks, some attention should be given to the width of the photopeak, and it should be recognized that width in MeV increases with increased energy.

A typical sequence of events leading to the qualitative identification of the components of a spectrum might consist of the following procedures:

1. Determine the location of photopeaks and convert to energy.
2. Compile a list of possibilities (*1*).
3. Eliminate from the list those radionuclides that have half-lives too short to be probable.
4. Look for radionuclides that appear more than once on the list (photopeaks at different energies).
5. Consider the possibility of parent-daughter relationships.
6. Check the decay schemes of those radionuclides from 4 above to see if any major photopeaks do not appear in the spectrum. (CAUTION: Consider the possibility that they may be hidden by some photopeak of slightly different energy.)
7. Compare the unknown spectrum against spectra of the possible choices for points of similarity (*2*).

The spectrum region in excess of 1 MeV is generally much less crowded by possibilities, and it is often helpful to examine the high-energy photopeaks first. The above process should reduce the list of possibilities to only a few radionuclides for any photopeak and in many cases may be sufficient for identification.

In the event that several possibilities still exist, an effort should be made to determine the half-life of the unknown radionuclide. This is accomplished most simply by recounting the sample several times and plotting a standard decay curve of the net counts under the photopeak as a function of time. To determine the net counts under the photopeak, allowance must be made for the influence of the Compton continuum resulting from higher energy peaks. A relatively simple method of doing this is to smooth the curve under the photopeaks and subtract this amount from the gross sum under the peak (fig. 5).

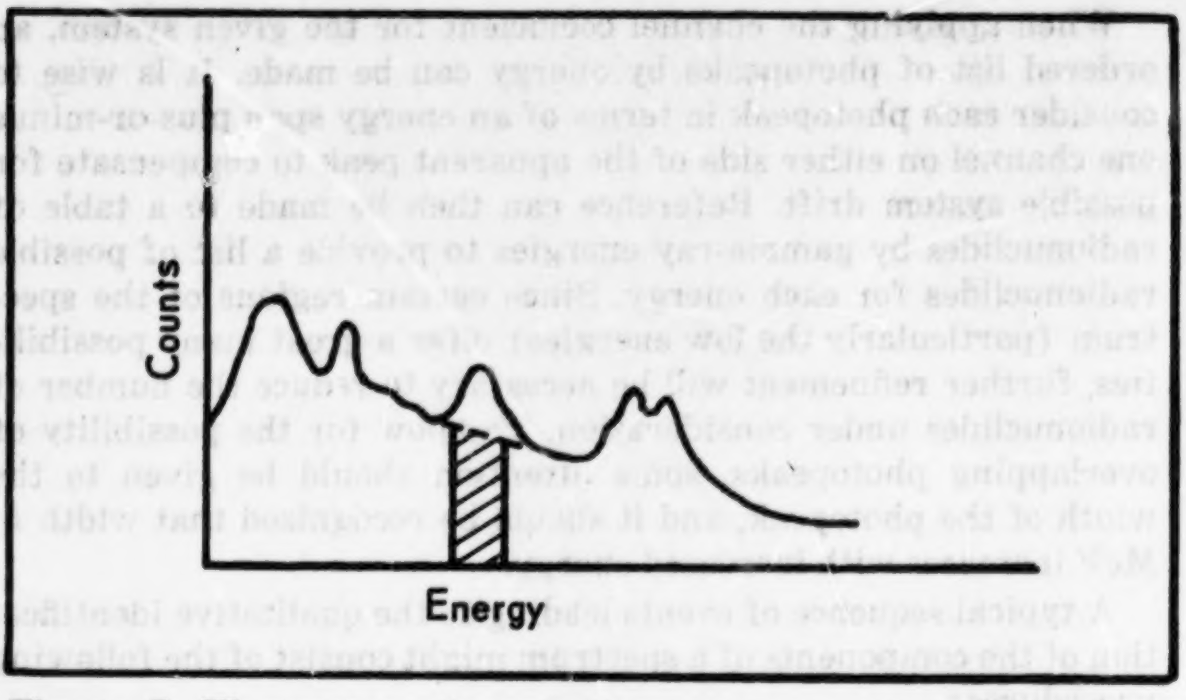


Figure 5. Illustration of correction of counts for influence of Compton continuum.

The analysis of the decay curve is accomplished by standard procedures. Complex decay curves are analyzed by the successive subtraction of the longer-lived components and replotting.

QUANTITATIVE ANALYSIS

The quantitative determination of the components of a gamma spectrum is the culmination of the efforts expended in the process of energy calibration, efficiency calibration, and qualitative identification. There are several methods of analysis that can be employed for manual computation of quantitative results.

APPROXIMATION BY COMPTON SUBTRACTION

As described briefly in the section on qualitative analysis, the net counts in a given photopeak can be approximated by smoothing the Compton continuum under the photopeak and subtracting the two. By applying an efficiency factor obtained by the same method, a disintegration rate can be computed. It may be found that the radionuclide in question has several photopeaks and one must be chosen for computation. The choice of the most appropriate photopeak involves consideration of the following:

1. The photopeaks must be free from overlapping peaks of other radionuclides to permit drawing of the Compton continuum.
2. It is desirable to utilize a peak having high-counting efficiency. The efficiency will be a function of energy (low energies are detected more efficiently) and gamma abundance.
3. Compton interference is reduced at higher energies.

This relatively simple method is the least accurate, but may be the only method in some areas. For routine environmental surveillance, more refined methods are recommended.

SPECTRUM STRIPPING

Most gamma spectra are not limited to a single radionuclide but contain photopeaks from several radionuclides. In order to quantitate each of these radionuclides, it is necessary to isolate the contributions from each source before applying efficiency corrections. Spectrum stripping is a method designed to minimize the effect of the Compton continuum on lower energy photopeaks by continually subtracting, or stripping, one radionuclide at a time, starting at the highest energy photopeak and working toward zero energy. In each successive subtraction, it is assumed that all of the counts (minus background) remaining in the highest energy photopeak are due to a single radionuclide. This is one weakness of the method.

The procedure for spectrum stripping might involve the following steps:

1. Identify the photopeaks and associated radionuclides.
2. Subtract background channel-by-channel.
3. Sum the counts within the highest energy photopeak.
4. Determine the ratio between the counts per minute of that photopeak and the counts per minute of a standard spectrum for a given radionuclide taken under identical conditions (that is, same shield, crystal, geometry, and so forth). This will be the coefficient by which each of the counts in each channel of the standard spectrum will be multiplied.

5. Perform a channel-by-channel subtraction of the adjusted standard spectrum (from 4 above) from the sample spectrum.

6. Repeat steps 3 through 5 using the highest energy photopeak that remains and the appropriate standard spectrum.

7. The activity of each radionuclide in the sample will be the product of the coefficient obtained in 4 above and the activity of the standard spectrum.

This method is a rather tedious process for manual calculation and has several limitations:

1. It is assumed that all of the counts in the highest energy photopeak that remain are due to a single radionuclide. This is not always true (consider iodine 131 and cesium 137 in a mixture).

2. Efficiency for detection of high-energy peaks is intrinsically lower than for lower energies.

3. Errors associated with the procedure tend to concentrate in the calculation of the lower energy radionuclides.

SIMULTANEOUS EQUATION (MATRIX) METHOD

A further refinement of the spectrum stripping procedure is a method that assumes a constant shape for the spectrum of any radionuclide, but is a method that can also compensate for overlapping or hidden photopeaks. In addition to the requirement of standard spectra, it is necessary to know which radionuclides are or may be present in the spectrum, and to have available a photopeak in a different energy region of the spectrum for each radionuclide. (Milk samples are particularly suited to this type of analysis since biological discrimination limits the available radionuclides.)

If a constant shape is assumed for the spectrum of a given radionuclide then any portion of that spectrum can be described in terms of the photopeak. This is the basic principle of this method.

For any one radionuclide (A) two basic parts can be considered: (1) the photopeak of interest will be called A_A and (2) the interference caused by A in any other spectral region (A_X). A_X is the interference coefficient of radionuclide A in region X and is found from the standard spectrum of A.

$$A_X = \frac{\text{Counts in X owing to A}}{\text{Counts in } A_A}.$$

Since the photopeak may contain counts that are due to other radionuclides, two further terms are introduced: (1) N_A equals net counts owing to A alone in the photopeak of A and (2) G_A equals the gross counts minus background in the photopeak used for A. If just a single radionuclide is present then $N_A = G_A$.

For the case of two radionuclides (A and B), using the same terminology, there may be mutual interference; that is, we may have a positive value for A_B and B_A. Then the net counts, because of A alone, in the photopeak of A will be given by: $N_A = G_A - B_A(N_B)$. In the same way, the net counts owing to B alone in the photopeak of B will be given by: $N_B = G_B - A_B(N_A)$. In these two equations, N_A and N_B are the only two unknowns. A_B and B_A (the interference coefficients) are found from the standard spectra of A and B, respectively, and G_A and G_B are found from the sample spectrum.

This process can be extended to additional radionuclides. For a milk sample containing iodine 131, cesium 137, barium-lanthanam 140, and potassium 40, the equation for iodine 131 would take the following form:

$$N_I = G_I - Cs_I(N_{Cs}) - Ba_I(N_{Ba}) - K_I(N_K).$$

The other three equations can be written in the same form. Having four equations and four unknowns, they can be solved simultaneously to eliminate N_X from the right-hand side and to replace it with the expressions containing nothing but constants and G_x.

$$N_A = K_1(G_A) - K_2(G_B) - K_3(G_C) - K_4(G_D).$$

Once this type of empirical formula has been obtained, it can be utilized for all samples containing the same radionuclides counted in the same geometry.

SINGLE-CHANNEL ANALYZERS

The principles of gamma-ray interaction with crystal detectors, which were described for multichannel analyzers, apply equally well to single-channel systems. The differences occur primarily in the electronic circuitry that is employed to store the pulses emitted by the phototube. The single-channel analyzer is simpler in design, and consequently considerably reduced in cost. (A single-channel analyzer is available for one-third to one-half the cost of a multichannel unit.)

Principle of Operation

Single-channel analyzers are equipped with discriminator circuitry that allows only pulses representing energies between E and E+ΔE to be recorded as counts at any given time. This is accomplished by permitting both the baseline (or minimum value of E) and the window width (or value of ΔE) to be preset by the operator. Both of these values are normally controlled by tenturn potentiometers. On some single-channel systems, the baseline discriminator may be motor driven, at the operators' discretion, to provide a differential scan of a wide energy band. In this case, the rate of sweep is constant and the end product is similar to that obtained by a multichannel analyzer. It should be noted that the counts per minute will be determined by the length of time each interval E+ΔE was examined and not by the total time of the scan. This type of scan is generally more valuable for qualitative rather than quantitative results.

Qualitative results can be most easily obtained by locking the discriminators on a given energy interval (a single photopeak from a given radionuclide). Calibration procedures for determining efficiencies are similar to those for multichannel analyzers. In addition, an energy calibration of photon energy versus baseline setting is also advisable. The exact location of the photopeak can be accomplished by using a source of the appropriate radionuclide. Some consideration should be given to the value used for ΔE when attempting quantitative analysis. In order to insure full coverage of the photopeak and to provide compensation against small gain shifts, it is generally advisable to use a window width that is equal to two times the full peak width at half-maximum peak height. This value will change with energy and should be determined for each radionuclide to be considered.

Although the usefulness of single-channel analyzers is limited when applied to environmental surveillance, there are some areas where they can be applied very successfully. They can be used as effluent monitors for gases or water released by operating reactors. In this case, a single radionuclide most indicative of the release of radioactivity by the reactor, either as a fission or corrosion (induced radioactivity) product, is selected and the window set to span a characteristic photopeak of that radionuclide.

Any changes in radioactivity levels at that energy can be readily detected by the use of a chart recorder showing radioactivity versus time. (This application may be either qualitative or quantitative.)

Tracer studies also provide a fertile field for the application of single-channel analyzers. For this purpose, radioactivity levels are fairly high, eliminating the need for long counting times, and a single radionuclide is generally used that eliminates the necessity of a preliminary qualitative analysis prior to quantification. Since radioactivity levels are high relative to background in this case, a minimum amount of shielding (or none in some cases) may be used, thus reducing the cost of the total system.

REFERENCES AND BIBLIOGRAPHY

(*1*) DERMATOSIAN, E. and M. MCKEOWN. Table of gamma rays emitted by radioactive nuclei, BNL–605 (T–177). Clearinghouse for Federal Scientific and Technical Information, Springleld, Va. 22151.

(*2*) HEATH, R. L. Scintillation spectrometry, IDO–16880, Vols. 1 and 2. Technical Information Division, U.S. Atomic Energy Commission, Washington, D.C.

(*3*) PUBLIC HEALTH SERVICE, DIVISION OF RADIOLOGICAL HEALTH. Radionuclide Analysis by Gamma Spectroscopy, Training Course Manual, Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio (September 1964).

PUBLIC HEALTH SERVICE, DIVISION OF RADIOLOGICAL HEALTH. Radiological Health Handbook. Clearinghouse for Federal Scientific and Technical Information, Springfield, Va. 22151 (Revised Edition September 1960).

CROUTHAMEL, C. E. Applied Gamma-Ray Spectrometry. The MacMillan Company, New York (1959).

PRICE, W. J. Nuclear Radiation Detection. McGraw-Hill Book Company, Inc., New York (1958).

CHASE, R. L. Nuclear Pulse Spectrometry. McGraw-Hill Book Company, Inc., New York (1961).

GLASSTONE, S. Sourcebook on Atomic Energy, 2d Edition. D. Van Nostrand Company, Inc., New Jersey (1958).

CROUTHAMEL, C. E., Editor. Applied Gamma-Ray Spectrometry. Pergamon Press, New York (1960).

SNELL, A. H., Editor. Nuclear Instruments and Their Uses. John Wiley and Sons, Inc., New York (1962).

Appendices to Chapter III

Typical specifications that might be used for shielding are included in the following pages. The background spectrum was obtained using a 4- by 4-inch NaI crystal in a steel shield similar to that specified.

APPENDIX A TO CHAPTER III

Typical Specifications for (Double Chamber) Shield

The low-background steel shield is to conform to the following specifications:

A. Double chamber assembly arranged so that both chambers share wall when assembled side by side with the base at floor level (app. B).

B. All walls shall be 6 inches of steel fabricated in 2-inch interlocking step construction plates.

C. Internal dimensions shall be 24 inches by 24 inches by 36 inches high.

D. Construction of double chamber unit shall be such that separation may yield two single shields with the purchase of additional plates to complete the construction of end walls at a later date.

E. Mounting of crystal assembly shall be on a slide-mounted shelf with the crystal assembly centrally located in the horizontal plane and the top of shelf 12 inches from the bottom of shield.

F. Slide assembly itself shall be mounted on supports located inside the shield and shall not be attached to the walls of the shield.

G. Both doors shall be hinged at outside of double assembly.

H. All material used in construction shall be free of radioactive contamination. For purposes of this specification, this shall be determined in the following manner: The assembled shields with 4- by 4-inch NaI(Tl) crystal assembly shall have a background that is lower than the attached spectrum (app. C) over the 0 to 2 MeV range or shall exceed this spectrum by no more than 10 percent. In either event, there shall be no areas in this spectrum in which an integrated area covering approximately 100 keV exceeds the average of the immediately adjacent 50 keV areas of higher and lower energy by more than 10 percent. The energy peak at 510 keV representing positron annihilation from cosmic secondaries will be exempted for purposes of this part. In general with

the one exception noted, the background must present a smooth continuum.

I. Listed as separate items, but included in the bid, will be the following options:

1. One-eighth-inch lead and $\frac{1}{16}$-inch cadmium liners for shields.
2. Sealing of chambers for purpose of feeding aged air to interior of shields. Facilities for introducing and venting this air shall also be included.
3. Installation at customer's site.

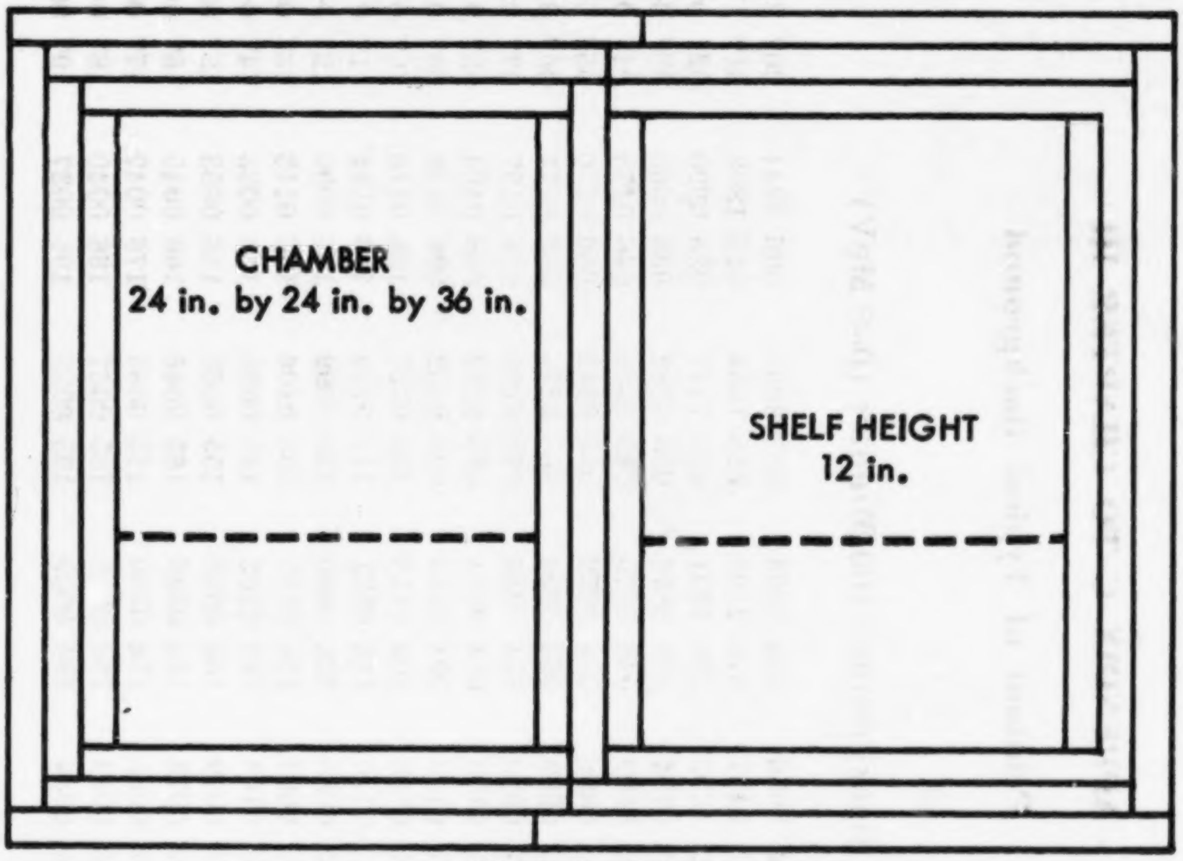
Shield shall be shipped disassembled, and the individual pieces will be of such dimensions as to pass through a 34-inch door.

Bidder will submit dimensioned sketch showing proposed shield and crystal carriage.

MANUFACTURER'S NAME________________________

BRAND____________________ MODEL NO.____________

APPENDIX B TO CHAPTER III


CHAMBER
24 in. by 24 in. by 36 in.
SHELF HEIGHT
12 in.

All Plates 2 in.

Low-background steel shield [a]

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[a] This sketch is a suggestion of general type only, and construction will not be held to this particular design. (For convenience in handling, two 1-inch plates could be substituted for the 2-inch plates.)

APPENDIX C TO CHAPTER III

Printout of Typical Background

Background: 100 Minutes (0–2 MeV)

```
000 0000    001 1023    002 1647    003 3956    004 3404    005 3090    006 2941    007 2922    008 2622    009 2452
010 2548    011 2365    012 2427    013 2417    014 2193    015 1964    016 1803    017 1734    018 1570    019 1547
020 1563    021 1449    022 1386    023 1392    024 1301    025 1147    026 0990    027 0953    028 0857    029 0891
030 0790    031 0812    032 0750    033 0754    034 0814    035 0682    036 0663    037 0603    038 0599    039 0603
040 0569    041 0533    042 0527    043 0509    044 0470    045 0503    046 0473    047 0507    048 0472    049 0494
050 0495    051 0447    052 0441    053 0396    054 0372    055 0342    056 0350    057 0321    058 0382    059 0356
060 0323    061 0303    062 0352    063 0299    064 0304    065 0275    066 0271    067 0261    068 0255    069 0261
070 0218    071 0216    072 0231    073 0229    074 0244    075 0198    076 0197    077 0217    078 0200    079 0179
080 0189    081 0212    082 0187    083 0179    084 0214    085 0183    086 0184    087 0160    088 0175    089 0168
090 0181    091 0153    092 0144    093 0142    094 0163    095 0152    096 0128    097 0143    098 0148    099 0124
100 0129    101 0145    102 0137    103 0118    104 0118    105 0127    106 0118    107 0123    108 0119    109 0120
110 0115    111 0117    112 0107    113 0116    114 0121    115 0119    116 0114    117 0111    113 0101    119 0107
120 0111    121 0087    122 0107    123 0085    124 0089    125 0099    126 0099    127 0091    128 0102    129 0089
130 0113    131 0093    132 0077    133 0094    134 0101    135 0106    136 0113    137 0093    138 0088    139 0086
140 0104    141 0113    142 0107    143 0109    144 0102    145 0095    146 0093    147 0090    148 0084    149 0090
150 0074    151 0076    152 0069    153 0079    154 0068    155 0068    156 0053    157 0064    158 0056    159 0063
160 0066    161 0060    162 0059    163 0072    164 0045    165 0045    166 0045    167 0050    168 0054    169 0057
170 0055    171 0047    172 0053    173 0061    174 0050    175 0063    176 0042    177 0052    178 0039    179 0047
180 0044    181 0048    182 0051    183 0044    184 0041    185 0037    186 0040    187 0045    188 0047    189 0037
190 0046    191 0036    192 0035    193 0044    194 0039    195 0033    196 0037    197 0029    198 0040    199 0041
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APPENDIX D TO CHAPTER III

Detectors

It is felt that the choice of detectors for gamma-spectrum analysis is sufficiently important to warrant more detailed consideration than has been previously given. Although only NaI crystals are considered here, other types of detectors, such as solid state for alpha and beta pulse-height analysis, and inorganic scintillators, such as CsI, are available for special applications. In most cases these detectors will be compatible with the pulse-height analyzers used for gamma-spectrum analysis.

Sodium iodide crystals are available as either (1) solid crystals in a wide range of sizes or (2) well crystals in which the diameter and depth of both the crystal and well are variable.

A. Solid crystals are generally more applicable to environmental surveillance than are well crystals, since they can accommodate much larger samples without going to very large and expensive units. For most applications a minimum detector size of 3 inches by 3 inches is recommended. (Many laboratories use 4- by 4-inch detectors.) This will permit more complete absorption of the higher energy photons with less chance for escape of degraded photons from Compton interactions. The net result is to reduce the height of the Compton continuum relative to the photopeak and increase system sensitivity. Because of the positioning of the sample on top of the crystal, it is unlikely that the detection efficiency for any energy gamma ray would exceed 20 percent (with the exception of point sources), regardless of geometry. The background will also increase as the volume of the detector increases so that the gain in sensitivity will be partially offset.

B. Well crystals are particularly useful for counting samples of very small volume. The size of the sample will depend on the volume of the well. Since the sample is physically located entirely within the detector, photons emitted in essentially all directions have a chance of interacting with the crystal. Much higher efficiencies are possible (for low energy photons this can be in excess of 50 percent). The introduction of a well into the crystal will reduce the amount of crystal material available for photon interaction and decrease the probability of total absorption of the photon. This increases the relative height of the Compton continuum and tends to decrease the detection efficiency. Generally for a ½- to 1-inch-diameter well, an overall diameter of 3 to 4 inches is recommended. Increasing the diameter of the well with a subsequent increase in crystal diameter is possible, but the cost of this type assembly is greatly increased.

The specifications for a NaI detector can be written to provide an integral unit in which crystal and photomultiplier tube are coupled within a sealed package. The advantages of this type assembly include better stability and freedom from drift because of temperature effects. The unit is hermetically sealed, eliminating the possibility of loss or movement of the optical coupling compound between the crystal and photomultiplier tube and providing better protection against moisture and dust. The major disadvantage of this type assembly lies in the fact that loss of either the crystal, because of cracking, or the photomultiplier tube usually results in loss of the whole assembly. The individual components usually cannot be removed and repackaged.

The alternative for specifying the above would be the purchase of a unit in which the crystal and photomultiplier tube are optically coupled, but not sealed in a single container. This may be the only type of package available for larger crystals (4 inches in diameter or greater). In all cases, it is strongly recommended that the crystal and photomultiplier tube be purchased from a single source of supply.

It is possible to obtain crystal assemblies having guaranteed performance specifications for a slight increase in price (less than $100). In addition to having low potassium content and guaranteed resolution, the stability of the assembly is also guaranteed. Long-term drift can be guaranteed to a maximum of 1 percent per day, and gain shift, because of increased count rate, can also be held to 1 percent. It is felt that this premium performance guarantee is well worth the nominal sum involved.

Errata

The enclosed revised pages 5 and 9 are to be inserted in PHS Publication No. 999–RH–23, "Routine Surveillance of Radioactivity Around Nuclear Facilities," December 1966, Interlaboratory Technical Advisory Committee Report No. 1 by the Subcommittee on Environmental Surveillance.

Fate of Radioactive Releases

The three principal avenues of radioactive releases to the environment are: (a) atmospheric releases, usually from elevated stacks, (b) liquid releases into surface streams or the ground, and (c) solid wastes, including structural material, equipment, paper, cloth and so forth, and liquids absorbed in concrete or other matrix. All of these solid wastes may be buried or stored in an innocuous state.

Table 2. Biological properties of some radionuclides (*15*)

Radionuclide and source	Critical organ	Fraction from G.I. tract to critical organ [a]	Effective half-life (days)
Fission products:			
$^{131}\mathrm{I}$	Thyroid	0.3	7.6
$^{133}\mathrm{I}$	Thyroid	0.3	0.9
$^{135}\mathrm{I}$	Thyroid	0.3	0.3
$^{132}\mathrm{Te}$	G.I. tract	([b])	([b])
$^{137}\mathrm{Cs}$	Total body	1.0	70
$^{89}\mathrm{Sr}$	Bone	0.2	50
$^{90}\mathrm{Sr}$	Bone	0.09	6.4×10^{3}
$^{140}\mathrm{Ba}$	Bone	0.03	11
$^{103}\mathrm{Ru}$	G.I. tract	([b])	([b])
$^{106}\mathrm{Ru}$	G.I. tract	([b])	([b])
Rare earths [c]	G.I. tract	([b])	([b])
Neutron activated:			
$^{3}\mathrm{H}$	Body tissue	1.0	12
$^{14}\mathrm{C}$	Fat	0.5	12
$^{24}\mathrm{Na}$	Total body	1.0	0.6
$^{32}\mathrm{P}$	Bone	0.37	14
$^{41}\mathrm{Ar}$	Lung	—	0.08
$^{55}\mathrm{Fe}$	Spleen	2×10^{-3}	390
$^{59}\mathrm{Fe}$	Spleen	2×10^{-3}	42
$^{60}\mathrm{Co}$	Total body	0.3	9.5
$^{65}\mathrm{Zn}$	Liver	0.035	66
Mixture [d]	G.I. tract	([b])	([b])
Naturally occurring:			
$^{226}\mathrm{Ra}$	Bone	0.04	1.6×10^{4}
$^{228}\mathrm{Th}$	Bone	7×10^{-5}	690
Transuranic elements:			
$^{239}\mathrm{Pu}$	Lung	—	[e] 365

[a] Assuming that the radionuclide occurs in soluble form.

[b] Only an extremely small fraction is absorbed into the body from the gastrointestinal (G.I.) tract. Therefore, the dose received by the G.I. tract during passage of the radionuclide is the limiting consideration.

[c] Rare earths $^{141}\mathrm{Ce}$, $^{144}\mathrm{Ce}$, $^{143}\mathrm{Pr}$, $^{147}\mathrm{Nd}$, $^{147}\mathrm{Pm}$, also $^{91}\mathrm{Y}$ and $^{95}\mathrm{Zr}$.

[d] A mixture of other activation products such as $^{51}\mathrm{Cr}$, $^{54}\mathrm{Mn}$, $^{56}\mathrm{Mn}$, $^{58}\mathrm{Co}$, $^{64}\mathrm{Cu}$, and $^{65}\mathrm{Ni}$ may be present.

[e] Reference *20*.

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plex and would normally involve an exorbitant number of sampling locations. A more realistic approach is to limit the locations along established perimeters of the plant and reduce the sampling and analytical load by gross screening and/or compositing of samples. As the average concentrations of particulate radionuclides approach limits of safety regulations, the size distribution of respirable particulates should be given consideration (*27–29*).

Sampling Equipment

Particulate samples are normally collected on a filter medium in conjunction with an air pump and a flow-measuring device. In this way, the sample is deposited in a uniformly thin layer which is suitable for direct alpha and beta measurement and for gamma spectrometry. For efficient measurement of alpha activity, porous collecting filters and heavy dust loadings should be avoided. The above samples can be further used in part or in whole as a source for subsequent individual or composite subsamples for radionuclide analysis, particle size studies, autoradiography, and the like.

Membrane filters of pore size 0.8 to 3 microns in diameter are the preferred media. They provide a fairly constant flow over a wide range of filter loadings.

The most satisfactory air movers are positive displacement pumps. Vane pumps are more suitable for use with filters when the head losses exceed 25 or 30 cm of mercury.

Selection of sampling equipment is dependent on the details of measurement procedures. For routine gross alpha and beta measurement with inexpensive scintillation or proportional counters (*30, 31*), samples of 30 to 70 m^3 on a filter area of 10 to 15 cm^2 are acceptable. However, since gamma-spectrometric analysis may also be required, a more useful sample would be 500 m^3 or more per day. Alternately, small samples may be utilized for the gross assays as above, with a larger sample of 2,000 m^3 available for gamma-spectrometric analysis. In this case, an 8- by 10-inch filter, such as the MSA 1106 BH glass fiber filter, is suggested (*32*). A glass fiber filter 3.5 inches in diameter is recommended when neither alpha assays nor gamma spectrometry is required. More detailed information on filter media characteristics is given in the appendix on page 25.

The surveillance of facilities from which nonparticulate radionuclides may be released requires special techniques. For the sampling of gaseous molecular iodine 131, an activated carbon canister placed between the filter and pump is recommended. For this, a canister satisfying U.S. Bureau of Mines Approval No. 2306 is suggested (*33*).

Each of the above sampling systems should be provided with a vacuum gauge and flow meter which, with the sampler, is mounted in

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